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SAN FERNANDO VALLEY STATE COLLEGE FOUNDATION

Northridge, California 91324

ANALYTICAL STUDIES PERTAINING TO THE MEASUREMENT OF LIGHT ABSORPTION IN AIR

Final Report

October 1968

Contract No. N00014-66-C0098

ARPA Order No. 306

This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research, and the Department of Defense.

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ABSTRACT

Experimental methods proposed for the measurement of the absorption coefficient of light in air are analyzed. The calculations are primarily intended for the wavelength regions where, for certain wavelengths, the absorption coefficient to be measured is estimated to be of the order of 10^{-9} cm^{-1}. The effects of scattering, heat conduction, static and acoustic pressure variations are examined and are related to the problem of detecting the signals produced by the absorption of heat from the laser beam. Both pulsed and cw schemes are considered. The measurement of the static pressure rise in a narrow tube appears as a feasible, although marginal method which offers some hope of success. An interferometer scheme also appears feasible. Finally, the linear absorption coefficient for the 10.6μ wavelength CO_2 laser beam has been measured in air.

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1. INTRODUCTION

Propagation of light at very high intensities produces changes in the properties of the medium through which the light propagates. These changes in the medium in turn affect the propagation of the beam. Such effects have been observed in liquids and gases and also take place in the atmosphere if laser beams of sufficiently high intensity were caused to pass.[a] The laser beam would be defocused by the refractive index gradient produced in the atmosphere by the absorption of energy from the beam. The work of Brueckner goes beyond this first order effect and treats the combined thermodynamic, mechanical, and optical instability resulting from the interdependence of density distribution, temperature and ray path. In this report concern is primarily with the linear absorption coefficient of the atmosphere for high intensity laser beams of various frequencies, and, in this section, with the first order effect of defocusing. Hence a review of the principles involved in determining beam spread is presented here.

When thermal energy is deposited in the atmosphere (assumed to be an ideal gas) by absorption from a laser beam, the resulting effects on pressure, temperature, density, and molecular velocity are determined by: (1) the ideal gas laws, (2) the equation of continuity, (3) Newton's second law of motion, and (4) conservation of energy. These effects are derived in many standard texts.[b] Applying the law of conservation of energy, one obtains the energy

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a. K.A. Brueckner: I.D.A. Laser Summer Study, 1963, or I.D.A. Research Paper P-42. Report on Laser Summer Study, The Inst. for Radiation Physics and Aerodynamics. U.C.S.D., Aug. 1965.

b. Knudsen & Katz: FLUID DYNAMICS AND HEAT TRANSFER, McGraw Hill, 1958.

balance equation for ideal gases (Eq. 2-55 of Knudsen and Katz):[b]

$$\rho C_p' \frac{dT}{dt} = K\nabla^2 T + \frac{dP}{dt} + q' + \Phi, \tag{1.1}$$

where ρ = mass density

T = absolute temperature

C_v' = heat capacity at constant volume

C_p' = heat capacity at constant pressure

t = time

P = pressure

γ = C_p'/C_v'

K = thermal conductivity

Φ = viscous dissipation function

q' = rate of heat generation in gas per unit volume.

Assuming K and Φ negligible, and

$q' = \alpha I$,

where α = linear absorption coefficient of the atmosphere (ideal gas) for the laser beam,

I = intensity (power per unit area) of the laser beam,

then equation (1.1) becomes,

$$\rho C_p' \frac{dT}{dt} - \frac{dP}{dt} = q' = \alpha I. \tag{1.2}$$

For an ideal gas

$$C_p' - C_v' = \frac{P}{\rho T}. \tag{1.3}$$

Hence,

$$C_v (\gamma - 1) \frac{dT}{dt} = \frac{1}{\rho}\frac{dP}{dt} - \frac{P}{\rho^2}\frac{d\rho}{dt}. \tag{1.4}$$

Substituting (1.4) into (1.2), one obtains a relationship independent of temperature as follows:

$$\rho C_p' \left\{ \frac{1}{(C_v'(\gamma-1)} \left[\frac{1}{\rho} \frac{dP}{dt} - \frac{P}{\rho^2} \frac{d\rho}{dt} \right] \right\} - \frac{dP}{dt} = \alpha I$$

$$\frac{1}{\gamma-1} \left(\gamma \frac{dP}{dt} - (\gamma-1) \frac{dP}{dt} - \frac{\gamma P}{\rho} \frac{d\rho}{dt}\right) = \alpha I \tag{1.5}$$

$$\frac{dP}{dt} - \frac{\gamma P}{\rho} \frac{d\rho}{dt} = (\gamma-1)\, \alpha I.$$

Now make use of a relationship for the speed of sound in a gas,

$$c_o^2 = \gamma P/\rho, \tag{1.6}$$

and one obtains:

$$\frac{d}{dt} (P - c_o^2 \rho) = (\gamma-1)\, \alpha I. \tag{1.7}$$

A concise treatment of the effects of density change on beam spread and instability has been developed by Brueckner.[a] His basic equations are obtainable from (1.7) by taking the Laplacian of both sides:

$$\frac{d}{dt} (\nabla^2 P - c_o^2 \nabla^2 \rho) = (\gamma-1)\, \nabla^2 I\alpha. \tag{1.8}$$

Now, using the equation of motion

$$-\underline{\nabla} P = \rho \frac{d\underline{v}}{dt}, \tag{1.9}$$

the equation of continuity,

$$-\frac{\partial \rho}{\partial t} = \underline{\nabla} \cdot (\rho \underline{v}), \tag{1.10}$$

along with the operator identity,

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \underline{v} \cdot \underline{\nabla}, \tag{1.11}$$

where $\underline{v}$ is the fluid velocity, one obtains an expression independent of pressure as follows:

$$\nabla^2 P = -\underline{\nabla}\cdot(\rho\frac{d\underline{v}}{dt}) = -\frac{d}{dt}(-\frac{\partial\rho}{\partial t}) + \underline{\nabla}\cdot(\underline{v}\frac{d\rho}{dt})$$

$$= \frac{\partial^2\rho}{\partial t^2} + \underline{v}\cdot\underline{\nabla}\frac{\partial\rho}{\partial t} + \underline{\nabla}\cdot[\underline{v}(\frac{\partial\rho}{\partial t} + \underline{v}\cdot\nabla\rho)] \qquad (1.12)$$

$$= \frac{\partial^2\rho}{\partial t^2} - \rho(\underline{\nabla}\cdot\underline{v})^2 - \underline{v}\cdot\underline{\nabla}[2\rho\underline{\nabla}\cdot\underline{v} + \underline{v}\cdot\underline{\nabla}\rho].$$

For small pressure and density fluctuations, neglecting gradients in $\underline{v}$, one can linearize equation (1.12) by the approximation,

$$\nabla^2 P = \frac{\partial^2\rho}{\partial t^2}.$$

Hence the equation for density change in a gas traversed by a laser beam is:

$$\frac{d}{dt}(\frac{\partial^2}{\partial t^2} - c_o^2\nabla^2)\rho = (\gamma-1)\nabla^2 I\alpha. \qquad (1.13)$$

If density changes occur slowly, so that

$$\frac{\partial^2\rho}{\partial t^2} << c_o^2\nabla^2\rho,$$

they may be approximated to occur at constant pressure, and equations (1.7) or (1.13) simplify to the following:

$$\frac{\partial\rho}{\partial t} = -\frac{\gamma-1}{c_o^2} I\alpha. \qquad (1.14)$$

Solutions of (1.13) and (1.14) combined with the eikonal equation have been obtained by Brueckner.[a] Suffice it here to obtain a very elementary solution to (1.14) to show how an initial first order effect of the spreading of an initially parallel laser beam depends upon α.

Before introducing geometrical optics, it may be helpful to arrive at equation (1.14) from even more elementary considerations. At constant pressure the heat per unit volume absorbed from the laser beam by an ideal gas is:

$$\frac{dQ}{V} = \alpha I \, dt = \frac{NC_p \, dT}{V} = \frac{\rho C_p \, dT}{M} \tag{1.15}$$

where N = number of moles

M = mass per mole

C_p = molar specific heat at constant pressure

C_v = molar specific heat at constant volume

V = volume.

Therefore,

$$\alpha I = \frac{C_p}{M} \rho \frac{dT}{dt} = - \frac{C_p}{M} T \frac{d\rho}{dt} = - \frac{c_o^2}{\gamma - 1} \frac{d\rho}{dt} \tag{1.16}$$

where use is made of the expression found in elementary texts for the speed of sound in a gas:

$$c_o^2 = \frac{\gamma R}{M} T = \frac{C_p}{C_v} (C_p - C_v) \frac{T}{M} = C_p(\gamma - 1) \frac{T}{M},$$

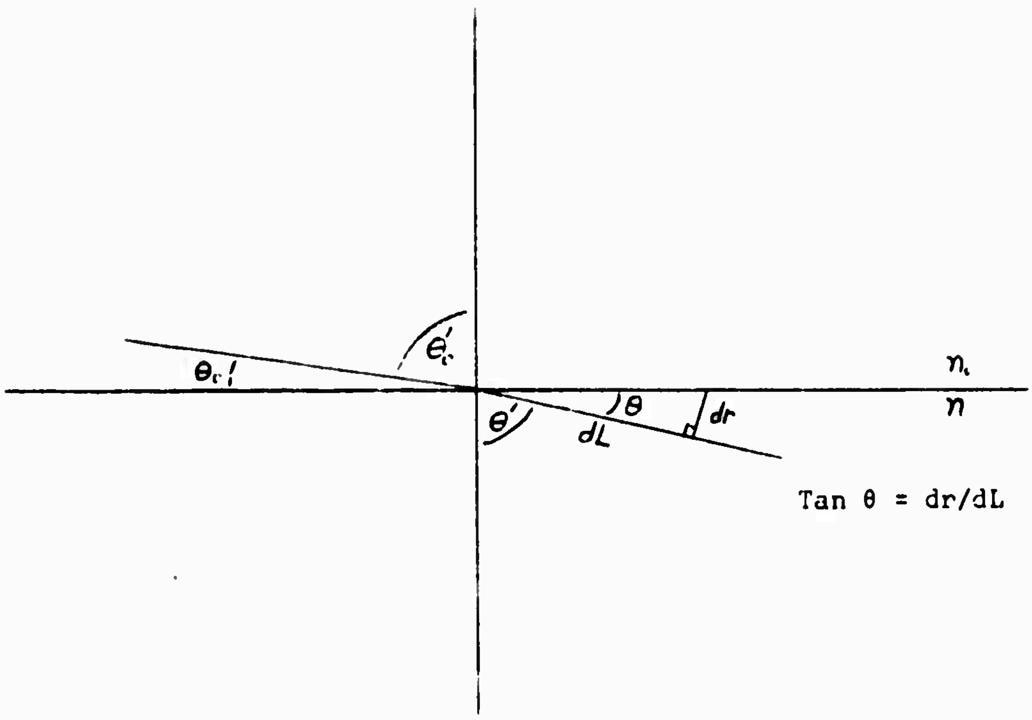
and, at constant pressure,

$$\frac{dT}{T} = - \frac{d\rho}{\rho} .$$

Equation (1.16) is another form of (1.14):

$$\frac{\partial \rho}{\partial t} = - \frac{\gamma - 1}{c_o^2} \alpha I. \tag{1.14}$$

The angular ray deflection may be approximated from Snell's law, where L is the coordinate in the direction of propagation, and r is a transverse coordinate, as illustrated in Figure 1.1.



$$n \sin \theta' = \text{constant} = n \cos \theta \qquad (1.17)$$

Figure 1.1. Ray Trace Illustrating Snell's Law

Differentiating Snell's law with respect to L, one obtains:

$$0 = - n \sin \theta \frac{\partial \theta}{\partial L} + \frac{\partial n}{\partial L} \cos \theta$$

or,

$$\frac{\partial \theta}{\partial L} = \frac{1}{n} \frac{\partial n}{\partial L} \cot \theta = \frac{1}{n} \frac{\partial n}{\partial L} \frac{dL}{dr} = \frac{1}{n} \frac{\partial n}{\partial r} . \quad (1.18)$$

Variations in refractive index of air may be closely approximated by equation (1.19):[c]

$$\delta n = \frac{\delta \rho}{\rho_o} (n_o - 1). \quad (1.19)$$

Hence (1.18) becomes, since $n \approx 1$,

$$\frac{\partial \theta}{\partial L} = \frac{1}{n} \frac{n_o - 1}{\rho_o} \frac{\partial \rho}{\partial r} \approx \frac{n_o - 1}{\rho_o} \frac{\partial \rho}{\partial r} . \quad (1.20)$$

Now equation (1.14) may be integrated and substituted into (1.20):

$$\rho - \rho_o = - \frac{\gamma - 1}{c_o^2} \alpha \bar{I} t,$$

where $\bar{I}$ is the time average intensity.

Hence,

$$\frac{\partial \theta}{\partial L} = - \frac{(n_o - 1)}{\rho_o} (\gamma - 1) \frac{\alpha t}{c_o^2} \frac{\partial \bar{I}}{\partial r} . \quad (1.21)$$

The change in intensity due to spreading of the beam may be approximated by:

$$\frac{\partial \bar{I}}{\partial r} = \frac{\partial}{\partial r} \bar{I}_o \frac{r_o^2}{[r(L)]^2} = - \frac{2 \bar{I}_o r_o^2}{[r(L)]^3} . \quad (1.22)$$

Hence equation (1.21) becomes

$$\frac{\partial \theta}{\partial L} = \frac{2(n_o - 1)(\gamma - 1) \alpha \bar{I}_o t \, r_o^2}{\rho_o \, c_o^2} \frac{1}{[r(L)]^3} = \frac{B}{[r(L)]^3} \quad (1.23)$$

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c. (Kuiper and Middlehurst: TELESCOPES, U. Chicago Press.)

where B is the constant so defined:

$$B = \frac{2(n_o-1)(\gamma-1)\alpha\bar{I}_o t\ r_o^2}{\rho_o\ c_o^2} \tag{1.24}$$

Equation (1.23) may be integrated easily, for a beam initially parallel, to determine the beam spread. From Fig. 1.1,

$$\tan\theta = \frac{dr}{dL} \approx \theta, \quad \text{or } dr \approx \theta\ dL.$$

Substituting these relationships into (1.23), one obtains:

$$d\theta = \frac{B}{r^3}\,dL = \frac{B}{r^3}\,\frac{dr}{\theta}\ . \tag{1.25}$$

Then, upon integration, one obtains:

$$\frac{1}{2}\,\theta^2 = \int_o^\theta \theta d\theta = B\int_{r_o}^{r} \frac{dr}{r^3} = \frac{B}{2}\,[\frac{1}{r_o^2} - \frac{1}{r^2}].$$

Hence,

$$\frac{dr}{dL} \approx \theta = \frac{B^{\frac{1}{2}}\sqrt{r^2-r_o^2}}{r\ r_o}\ ,$$

which may be integrated to give:

$$\sqrt{r^2-r_o^2} = \int_{r_o}^{r} \frac{rdr}{\sqrt{r^2-r_o^2}} = \frac{B^{\frac{1}{2}}}{r_o}\int_o^L dL = \frac{B^{\frac{1}{2}}\ L}{r_o}$$

Hence it follows that the beam radius increases with distance according to to:

$$r = r_o\,[1 + \frac{BL^2}{r_o^4}]^{\frac{1}{2}} \tag{1.26}$$

and the beam deflection or angle of spread is:

$$\theta = \int_o^\theta d\theta = \int_o^L \frac{B}{r_o^3}\,dL = \frac{r_o^3}{B^{\frac{1}{2}}}\int_o^L \frac{dL}{\left[L^2 + \frac{r_o^4}{B}\right]^{3/2}} = \frac{B^{\frac{1}{2}}}{r_o}\,\frac{L}{\left[L^2 + \frac{r_o^4}{B}\right]^{\frac{1}{2}}}$$

or

$$\theta = \frac{BL}{r_o^3}\,[1 + \frac{BL^2}{r_o^4}]^{-\frac{1}{2}} \tag{1.27}$$

Hence it is seen that to a first approximation the beam spread is proportional to $B^{\frac{1}{2}}$, and B is proportional to beam power, time, and α, the linear absorption coefficient. It is therefore important that methods be available to measure values of α for various laser frequencies where high intensities are physically realizable.

2a. Statement of the Purpose of the Report.

It is of interest to know the amount of heat evolved in a material through which a laser beam is passing. A parallel beam of light is attenuated as it passes through matter and this attenuation is mainly attributed to two effects: scattering and absorption. Scattering on material particles results in the directional redistribution of the energy of the incident electromagnetic wave. Absorption results in the conversion of electromagnetic energy into other forms of energy. The energy lost by the electromagnetic field is ultimately converted into heat. This means that, possibly through the intervention of processes of atomic and molecular excitation, the energy abstracted from the electromagnetic field is redistributed among the available degrees of freedom of the material (gas), so that it is ultimately undistinguishable from energy obtainable from contact with a heat reservoir. We shall assume that the atomic and molecular processes responsible for the redistribution of energy among the available degrees of freedom take place so fast that at the scale at which we contemplate our observations the conversion of the absorbed energy into heat may be regarded as instantaneous. It should be noted that we are talking here about energy redistribution locally and not about its redistribution in space. The latter generally proceeds at a much slower rate. We shall thus proceed under the assumption that the energy absorbed from the electromagnetic field instantly appears at the site of absorption in the form of heat. Such an assumption is probably valid if our time resoltuion is no finer than 10^{-7} sec.

The absorption of light in solids and liquids is usually significant enough so that a direct measurement of the absorption is not difficult. In

gases, such as pure air, the measurement of the absorption of light is very difficult, because the rate of absorption is small and in the visible region it is completely masked by the much larger effect of scattering. The situation is somewhat more favorable in the infrared beyond 1.2 microns.

The optical effects of scattering and absorption combine so that the intensity of a plane wave passing through matter is described by the equation

$$I(x) = I_0 e^{-\beta x} , \tag{2.1}$$

where the attenuation coefficient β is the sum of two terms:

$$\beta = \alpha + \sigma . \tag{2.2}$$

Here α is the absorption coefficient, σ the scattering coefficient and x is the direction of propagation.

In the spectral region where the ratio σ/α is large, it is not feasible to determine α by measuring the attenuation of a light beam through matter. The value of α must thus be determined not by the change produced in the radiation but by the change produced in the absorbing matter. We already noted that the absorption of radiation results in an increase in the thermal energy of the absorbing matter, therefore we shall address ourselves to problems pertinent to the detection and measurement of small quantities of thermal energy deposited in the path of a narrow light beam of the type that may be obtained from a laser.

It has been estimated that for certain wavelengths in and around the visible region of the spectrum α is of the order of 10^{-9} cm^{-1}. Scattering in pure air, as calculated from Rayleigh's theory, produces a much higher attenuation. For green light (0.5 microns), $\sigma = 1.7 \times 10^{-7}/cm$ for air free of all contaminants. We reserve the quantitative data for the next section, but we note here, that around 10 microns the situation is quite different.

First, significant and spectrally selective absorption is likely to be encountered because of the presence of the molecular bands of the constituents of air in this region. Second, Rayleigh scattering on molecules, which varies with the fourth power of the wavelength, is reduced to negligible values around 10 microns.

This report contains calculations pertaining to thermal and acoustic effects expected as a result of the passage of a powerful laser beam through air. The configurations examined are such which are likely to be chosen in experiments aimed at the determination of α in the 0.5 to 1.2 micron spectral region. We have included here the analysis of the pressure change in a completely enclosed cylinder whose walls are kept at a constant temperature, the calculation of the rate of dissipation of heat around a laser beam by means of thermal conduction, the generation of a quasi-static pressure increment, and an acoustic pulse by a laser pulse and some problems pertaining to the detection of an acoustic pulse or a sequence of acoustic pulses. The material as assembled here does not represent a complete investigation. It is a group of exploratory calculations preliminary to the experimental determination of the absorption coefficient of light in air.

2b. Relevant Properties of Air.

In this section we have collected such physical data concerning air that seem relevant for computations in later sections, or that may be needed in evaluating an experimental scheme proposed for the measurement of the absorption coefficient α.

Atmospheric air free of dust and other contaminations is a mixture of gases, primarily N_2, O_2, Ar, CO_2 and water vapor. Other gases are present in

such small proportions that their presence does not noticeably affect the physical properties of air that are of concern to us. Of the gases listed here only the proportion of water vapor is variable.

In what immediately follows we shall assume that we are dealing with clean, dry atmospheric air. By this substance we mean a gas mixture from which all water vapor has been removed and which contains the gases N_2, O_2, Ar and CO_2 in the proportion as they are universally found in the atmosphere[1]. We note in particular that such air contains 0.033 percent CO_2 by volume. Although this proportion seems small, it is well to remember that a relatively small amount of CO_2 or water vapor significantly affects the optical properties of air.

It is customary to specify the physical properties of gases at standard temperature and pressure; i.e. at 0°C and 760 torr. At these conditions the properties of dry atmospheric air relevant to this study are as follows:

Density: $\rho_o = 1.293 \times 10^{-3}$ g cm^{-3},

Specific heats: $c_p = 0.240$, $c_v = 0.172$ cal g^{-1} °C^{-1},

Sound velocity: $c_o = 3.31 \times 10^4$ cm sec^{-1},

Heat conductivity: $K = 5.68 \times 10^{-5}$ cal cm^{-1} sec^{-1} °C^{-1},

Index of refraction for $\lambda = 5000$ Å: $n = 1.000294$.

At standard temperature and pressure the particle density of every ideal gas is $L = 2.69 \times 10^{19}$ cm^{-3}. (Loschmid's number.) The variation of $n - 1$ with wavelength (dispersion) has been studied and is tabulated for a temperature of 15°C, which must be a convenient laboratory temperature in Sweden. The following data applicable at 15°C are obtained from the work of Edlén:[2]

1. Handbook of Chemistry and Physics, 46th ed. p. F-116.

2. B. Edlén, The dispersion of standard air, J. Opt. Soc. Amer. 43, 339, 1953.

λ(Angstroms)	5000	6000	7000	8000	9000	10000
$(n - 1) \times 10^4$	2.78	2.76	2.75	2.75	2.74	2.74

At the more comfortable laboratory temperature of 20°C the following data are applicable for 760 torr pressure:

Density: $\rho_{20} = 1.205$ g cm^{-3},

Sound velocity: $c_{20} = 3.44 \times 10^4$ cm sec^{-1},

Loschmid's number $L = 2.51 \times 10^{19}$ cm^{-3},

The value of $n - 1$ varies from 2.73×10^{-4} at 5000 Å to 2.69×10^{-4} at 10000 Å.

The attenuation of light caused by scattering on the molecules of air is calculable for Rayleigh's formula[3]

$$\sigma = \frac{32\pi^3(n-1)^2}{3L\lambda^4} . \qquad (2.3)$$

At standard temperature and pressure we obtain the following values of σ:

λ(Angstroms)	5000	6000	7000	8000	9000	10000
$\sigma \times 10^7$(cm^{-1})	1.70	0.81	0.44	0.25	0.16	0.10

Both (n-1) and L are proportional to ρ, therefore σ is also proportional to ρ. At constant pressure σ is inversely proportional to the absolute temperature. Therefore the values of σ at t = 20°C and 760 torr are about 7 percent smaller than the above tabulated values.

3. Absorption Experiment With a Steady Laser Beam.

We shall analyze the problems involved in the measurement of α by means of the following experiment: A certain volume of air is confined at a known

3. J.W. Strutt, Lord Rayleigh, On the transmission of light through an atmosphere, Phil. Mag. 47, 374, 1899; Scient. Papers 4, 397.

initial pressure. A laser beam passes through the confined volume and heats the enclosed air at a rate proportional to α. In a steady state, the heat losses of the system balance the heat gained from the laser. The increase of the static pressure within the vessel is measured and is used for computing the rate at which heat is added to the air.

The simplest experimental embodiment of this idea consists of a cylindrical tube kept at a constant temperature. The laser beam passes along the axis of the cylinder heating a smaller cylindrical volume of air concentric with the confining tube. A steady state is established when the rate at which heat is conducted to the walls is equal to the rate at which heat is absorbed from the laser beam.

In order that a technique of this type yield a valid measurement of α, certain physical conditions must be satisfied which are not easy to achieve experimentally. First, the confining tube must be long enough so that special thermal effects at the ends of the tube do not significantly affect the pressure of the confined air. Laser light is brought in through solid windows whose density is about 1000 times that of the confined gas. It is not unreasonable to assume that absorption per unit volume in the windows will be about 1000 times as great as absorption in the gas. Second, for $\lambda < 1.2\mu$ more laser light will be scattered than directly absorbed. The energy of the scattered radiation must be removed, it must not be allowed to add to the thermal energy of the confined gas. This removal could be accomplished, for example, by making the inner surface of the tube black and by making the tube of a material with good thermal conductivity.

Assuming that these experimental difficulties are mastered, we calculate the pressure increase dp attained in the vessel in the steady state as a result of energy being absorbed from the laser beam at a constant rate and conducted away through the air to the walls of the vessel which are kept at a constant temperature T_o. We assume a uniform circular laser beam of radius a in a vessel of radius b. Let the power incident in the laser beam be W, then the intensity of the beam is $I = W/a^2\pi$. Heat is deposited in the unit volume of the gas at the rate αJI, where J = 0.239 cal/joule.

Once the steady state is reached, no mechanical work is done. Therefore the heat balance equation is obtained by equating the rate at which energy is absorbed to the rate at which it is conducted away. (It is assumed that convection is neglected.) The rate of heat generation per unit volume is

$$q'(r) = \alpha JI \text{ when } r < a,$$
$$= 0 \text{ when } r > a.$$

The heat balance equation for a volume V is then

$$\int_V q'dv = - K \oint_S \nabla T \cdot \underline{n} dS, \tag{3.1}$$

where dS is the surface element, $\underline{n}$ is the unit normal to the surface, and K the thermal conductivity. It follows then in a well-known manner that

$$\nabla^2 T = - q'/K. \tag{3.2}$$

Since the only relevant coordinate in this problem is r, the distance from the cylinder axis, we have

$$\frac{1}{r}\frac{d}{dr} r \frac{dT}{dr} = - \frac{q'(r)}{K} \tag{3.3}$$

This differential equation is to be solved with the boundary condition $T = T_o$ for $r = b$ and the solution is required to remain finite for $r < b$. Although the solution is easily found, we do not need its explicit form because we are only interested in the pressure rise in the vessel and this is proportional to the increase of the average temperature.

In place of T we introduce the dimensionless variable

$$v(r) = \frac{K}{\alpha JI}(T - T_o). \tag{3.4}$$

Equation (3.3) is then replaced by

$$\frac{1}{r}\frac{d}{dr} r \frac{dv}{dr} = - g(r), \tag{3.5}$$

where $g(r) = 1$ for $r < a$ and 0 for $r > a$. The boundary condition is $v(b) = 0$. Then we obtain by integration of (3.5)

$$r \frac{dv}{dr} = - \int_o^r g(r')r'dr' = - f(r), \tag{3.6}$$

where $f(r) = \frac{1}{2} r^2$ for $r < a$, and $f(r) = \frac{1}{2} a^2$ for $r > a$.

The average increase in gas temperature over the entire vessel is proportional to

$$\bar{v} = \frac{1}{V}\int_o^b \int_o^{2\pi} \int_o^L v(r)rdrd\theta dz = \frac{b}{b^2}\int_o^b v(r)rdr. \tag{3.7}$$

Integrating by parts and noting that $v(b) = 0$, we get

$$\int_o^b v(r)rdr = -\frac{1}{2}\int_o^b r^2 \frac{dv}{dr} dr = \frac{1}{2}\int_o^b f(r)rdr.$$

Since $\int_o^b f(r)rdr = \int_o^a \frac{1}{2} r^3 dr + \int_a^b \frac{1}{2} a\, rdr = \frac{a^2b^2}{8}[2 - \frac{a^2}{b^2}]$, we have

$$\bar{v} = \frac{a^2}{4}[1 - \frac{a^2}{2b^2}]. \tag{3.8}$$

Therefore the average increase of temperature in the vessel is

$$dT = \frac{\alpha J I a^2}{4K} [1 - \frac{a^2}{2b^2}]. \tag{3.9}$$

The final expression for the increase of pressure in the chamber is obtained from (3.9) by using the gas law and by reintroducing the incident power $W = a^2 \pi I$.

$$dp = \frac{p_o}{T_o} \frac{\alpha J W}{4\pi K} (1 - \frac{a^2}{2b^2}). \tag{3.10}$$

Ordinarily the expression in the parentheses is nearly equal to one. Assuming a laser with 100 watts useful output, an initial temperature $T_o = 300°K$ and $\alpha = 10^{-9}$, the expression in front of the parentheses is 1.1×10^{-7} atmospheres. A pressure change of this order of magnitude should be detectable provided one can keep the temperature of the walls constant to within 0.3×10^{-4} °C. It might be quite difficult to accomplish this for a tube as long as 1 meter.

4. Dissipation of Heat in Air by Conduction.

In preparation for the discussion of experiments involving the heating of air columns by means of short laser pulses we shall calculate the rate at which thermal energy initially communicated to an air column is dissipated by thermal conduction alone.

The calculation is simplified by neglecting for the purpose of this calculation the aerodynamic properties of air. Thus we shall be concerned with a solid substance characterized by a heat conductivity K, density ρ,

and specific heat c_v.

It is shown in textbooks[4] that the temperature $v(x,y,z,t)$ satisfies the differential equation

$$\nabla^2 v - \frac{1}{\kappa}\frac{\partial v}{\partial t} = 0, \qquad (4.1)$$

provided that there are no sources or sinks of heat in the region under consideration. Here $\kappa = K/\rho c_v$ is the diffusivity of the substance.

We will assume a geometry of cylindrical symmetry and calculate the cooling of an infinite cylinder of radius a, whose temperature at the time $t = 0$ is one degree above the temperature of its surroundings. All space is assumed to consist of the same material characterized by the constant κ. The problem then requires the use of only one spatial variable, r, the cylindrical polar coordinate. The differential equation becomes

$$\frac{\partial v}{\partial t} = \kappa\left(\frac{\partial^2 v}{\partial r^2} + \frac{1}{r}\frac{\partial v}{\partial r}\right), \qquad (4.2)$$

and the initial condition is $v(r,0) = f(r)$, where

$$\begin{aligned} f(r) &= 1 \quad \text{for} \quad r < a, \\ &= 0 \quad \text{for} \quad r > a. \end{aligned}$$

As long as no walls are present, no boundary conditions are required. This differential equation is readily solved by the Hankel transformation technique[5] and the following result is obtained:

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4. H.S. Carslaw and J.C. Jaeger, Conduction of Heat in Solids, Oxford, 1959, 2nd ed. p. 9.

5. Ref. 4, p. 460.

$$v(r,t) = \int_0^\infty F_o(\sigma)\sigma J_o(\sigma r)e^{-\kappa\sigma^2 t}d\sigma, \tag{4.3}$$

where

$$F_o(\sigma) = \int_0^\infty rJ_o(\sigma r)f(r)dr. \tag{4.4}$$

In the present case

$$F_o(\sigma) = \int_0^a rJ_o(\sigma r)dr = \frac{a}{\sigma} J_1(\sigma a). \tag{4.5}$$

Hence the expression for the temperature distribution is

$$v(r,t) = a \int_0^\infty J_1(\sigma a)J_o(\sigma r)e^{-\kappa\sigma^2 t}\, d\sigma. \tag{4.6}$$

It is interesting to calculate the variation of temperature at the axis. From (4.6) we have for $r = 0$

$$v(0,t) = a \int_0^\infty J_o(\sigma a)e^{-\kappa\sigma^2 t}\, d\sigma. \tag{4.7}$$

This expression is changed by integration by parts into

$$v(0,t) = 1 - 2\kappa t \int_0^\infty J_o(\sigma a)\sigma e^{-\kappa\sigma^2 t}\, d\sigma. \tag{4.8}$$

From the standard formula[6]

$$\int_0^\infty J_o(\sigma a)e^{-p^2\sigma^2}\, \sigma d\sigma = \frac{1}{2p^2}\, e^{-\frac{a^2}{4p^2}} \tag{4.9}$$

we get

$$v(0,t) = 1 - e^{-\frac{a^2}{4\kappa t}}. \tag{4.10}$$

6. G.N. Watson, Theory of Bessel Functions, Cambridge, University Press, 1958, p. 393.

Thus the temperature at the center line decreases with t according to the formula $1 - e^{-\tau/t}$, where $\tau = a^2/4\kappa$. The quantity τ has the dimension of time. It is the time during which the temperature decreases by the amount 1/e. Although we are not dealing here with a simple exponential decay of temperature, it is still reasonable to regard τ as a thermal relaxation time. It is proportional to the cross sectional area of the beam.

Numerical computation of the thermal relaxation time requires the knowledge of κ for air in the applicable temperature range. This quantity entered first in equation (4.1) which is taken from the theory of heat conduction applicable to solids. When this theory is applied to a gas, the question arises what kind of specific heat is to be used in the equation

$$\kappa = \frac{K}{\rho c} \tag{4.11}$$

that serves as a definition of κ^2. Since we are contemplating a steady state situation in which no mechanical motion takes place, no work is being done and the entire heat energy that enters a volume of gas is converted into internal energy. The specific heat at constant volume is the one which is applicable to such a situation. For air at standard pressure and temperature, $\kappa = 0.255$ cm^2 sec^{-1}. Although κ depends on the temperature, this value of κ is probably a fair approximation to the correct value applicable at 20°C. With the value $\kappa = 0.255$ cm^2 sec^{-1} we obtain the following approximate values of relaxation times and associated frequencies applicable to beams of convenient cross sections.

Beam diam. (2a) mm	0.4	0.6	0.8	1.0	1.2	1.4	1.6	1.8	2.0
Relax. time (τ) msec	0.392	0.882	1.57	2.45	3.53	4.80	6.27	7.94	9.80
Frequency (τ^{-1}) Hz	2550	1134	637	408	283	208	160	126	10[illegible]

In a dynamic situation, when heat is added to a column of gas, not only heat conduction takes place but the gas is set in motion. This transient phenomenon was not taken into account so far. In what follows later, the dynamic phenomenon is of great importance and the results of this section are used for guidance concerning the situations when the effect of heat conduction may be neglected. Clearly one can only justify neglecting heat conduction if the dynamic phenomenon (oscillation) occurs so fast that only negligible cooling takes place during one cycle. Such will be the case when the frequency of the sound wave generated exceeds the appropriate characteristic frequency $\tau^{-1} = 4\kappa/a^2$.

When the air column is not free, as heretofore assumed, but confined in a pipe kept at a constant temperature, the method of calculation is different. The differential equation (4.1) is applicable in this case, but a boundary condition, absent in the case of the free air column, must be added. For a cylindrical vessel of inner radius b, the boundary condition is $v(b,t) = 0$, provided that the vessel is kept at a constant temperature T_0. (Here v denotes the excess over this temperature.) The solution of the heat conduction problem for a domain with a boundary is well known[7]. It is obtained by expanding the function to be determined in terms of the eigenfunctions of the differential equation (4.1) with the given boundary conditions. In the case of heat conduction within a cylinder of radius b the result is

7. Ref. 4, pp. 198-199.

$$v(r,t) = \sum_{n=1}^{\infty} A_n J_o(\alpha_n r) e^{-\kappa\alpha_n^2 t}, \tag{4.12}$$

where the numbers α_n are determined from the boundary condition $J_o(\alpha_n b) = 0$, and the coefficients A_n are calculated from the expansion

$$f(r) = \sum_{n=1}^{\infty} A_n J_o(\alpha_n r), \tag{4.13}$$

which is the explicit form of the initial condition. The A_n's are determined by the routine procedure applicable to expansions according to orthogonal functions. Their values depend on a and b. In the general case the value of A_n does not drop off rapidly with increasing n. We must therefore take into account a large number of terms in (4.12). Each term decays in time according to a different exponential rate $\kappa\alpha_n^2$. The time constants associated with these modes are $\tau_n = (\kappa\alpha_n^2)^{-1}$, and α_n is determined from the equation

$$\alpha_n b = \chi_n, \tag{4.14}$$

where χ_n is the n-th root of the equation $J_o(\chi) = 0$. The values of the first five χ's are: 2.405, 5.520, 8.654, 11.792 and 14.913.[8] The time constants are

$$\tau_n = \frac{b^2}{\kappa\chi_n^2}. \tag{4.15}$$

We note that the role of the τ_n's is not the same as that of τ in the case of the free cylinder because in the present case the time dependence of a mode is of the form $\exp(-t/\tau_n)$ and not $1 - \exp(-\tau/t)$.

By means of a physical argument we can readily infer that the confined

8. A. Gray and G.B. Mathews, Bessel Functions, 2nd ed., Dover Publ., New York 1966, p. 300.

cylinder will cool more rapidly than the free cylinder and that the relaxation time calculated for the free cylinder will be an approximation of the effective relaxation time of a confined cylinder for $b >> a$.

5. Pressure Rise in a Tube Caused by a Single Laser Pulse.

One might attempt to confine air in a cylindrical tube, shoot a laser beam through and measure the rise in pressure in the ube. The expected increase in pressure is proportional to the energy deposited in the air by the laser beam. Hence one expects that the absorption coefficient α might be measured in such an experiment.

It must be remembered that the walls of the tube holding the air are solid and have a much larger heat capacitance than the confined air. So eventually as thermal equilibrium is approached, the thermal energy deposited in the air is conducted to the walls, temperature and pressure in the vessel return to their values at the beginning of the experiment. The approach to thermal equilibrium may be thought of as consisting of a fast process of pressure equalization followed by a slower process of temperature equalization by heat conduction. The pressure equalization may be characterized by the time constant $2b/c$ where b is the radius of the tube and c the velocity of the sound. The rate at which heat conduction leads to thermal equilibrium must be calculated in the manner described in Section 4. The time constant $\tau = b^2/4\kappa$ certainly reveals the correct order of magnitude. The measurement of pressure increase should take place after the elapse of the time $2b/c$ following the passage of the laser beam but considerably before the elapse of the time τ. For a tube of 1 cm radius, $2b/c = 6 \times 10^{-5}$ sec and $\tau = 1$ sec.

The laser beam will generally be much narrower than the tube. Let the radius of the beam cross section be a, and let us assume that we are dealing with a very short laser pulse. Then at the end of the pulse we have an air cylinder of radius a within which the pressure has been raised to p + dp and the temperature to T + dT, while outside this cylinder the pressure is p and the temperature is T. According to the standard equation of heat balance

$$c_v \rho_o a^2 \pi dT = \alpha JQ, \tag{5.1}$$

where Q is the energy of the laser pulse in joules, J is the conversion factor 0.239 cal/joule, and ρ_o is the density of the air. From the ideal gas law and equation (5.1), we get

$$\frac{dp}{p} = \frac{\alpha JQ}{c_v \rho_o a^2 \pi T} . \tag{5.2}$$

On substituting the appropriate physical constants of air, we get for a = 0.1 cm and T = 300°K:

$$\frac{dp}{p} = 1.14 \times 10^2 \ \alpha Q. \tag{5.3}$$

Thus a laser pulse of 1 joule and an $\alpha = 10^{-9}$ cm^{-1} would give a relative pressure increase of one part in ten million. It seems then that, using a powerful laser of several joules output, an α of the order of 10^{-9} cm^{-1} would be just barely detectable. This would be the case if the entire confined space would be within the beam. Actually the beam diameter must be considerably smaller than the diameter of the tube that keeps the air confined. Consequently the pressure rise observable in the tube is much less than the pressure rise in the cylinder swept out by the beam.

An estimate of the pressure rise anticipated in the tube may be obtained by ascribing the entire gas in the tube some sort of average temperature $\overline{T}$. The heat balance equation (5.1) is then replaced by

$$c_v \rho_o b^2 \pi d\overline{T} = \alpha JQ. \tag{5.4}$$

This equation differs from (5.1) insofar as b^2 takes now the place of a^2. Consequently the pressure rise calculated from (5.4) will be reduced in proportion to the ratio of the cross sectional ratio of the beam to the cross sectional area of the tube. If, for example, $a = 0.1$ cm and $b = 1.0$ cm the value of dp/p is reduced by a factor of 100, thus driving it completely beyond reach for measurement.

It could be objected that the above estimate of dp/p is based on a dubious assumption of an average temperature and the application of thermodynamics to non-equilibrium states. The deduction may be replaced by the following one: After irradiation, the pressure of the air within the beam is p_1, its volume is v_1. In the rest of the vessel the pressure is p_o. The volume of the rest is v_2. Both volumes change during the process of pressure equalization which results in a final pressure P. Assuming no heat exchange,

$$p_1^{1/\gamma}\, v_1 = P^{1/\gamma}\, v_1' \quad \text{and} \quad p_o^{1/\gamma}\, v_2 = P^{1/\gamma}\, v_2'.$$

Moreover, the total volume does not change, therefore $v_1' + v_2' = v_1 + v_2$. Hence

$$P^{1/\gamma} = \frac{p_1^{1/\gamma}\, v_1 + p_o^{1/\gamma}\, v_2}{v_1 + v_2}\,, \tag{5.5}$$

and

$$P^{1/\gamma} - p_o^{1/\gamma} = \frac{v_1}{v_1 + v_2}\,(p_1^{1/\gamma} - p_o^{1/\gamma}). \tag{5.6}$$

From this last equation it also follows that in first approximation

$$P - p_o \sim \frac{v_1}{v_1 + v_2} (p_1 - p_o). \tag{5.7}$$

6. Generation of Sound by a Laser Pulse.

A laser pulse is sent through a gas which has a small absorption coefficient for the radiation sent through it. As a consequence of absorption a column of gas is warmed and the surrounding gas is set in motion. It is desired to determine the sound energy generated by the pulse and to determine the spectral distribution of the sound.

The solution of the problem is expected to contain the physical parameters of the gas which normally determine the velocity of propagation of sound in the gas, the rate at which radiated energy is absorbed in the gas, the length of the laser pulse and the cross-section of the laser beam.

It will be assumed that the gas is an ideal gas, that the laser beam is of circular cross-section, that the rate of energy dissipation and the pulse length are such that the resulting temperature, pressure and density changes may be treated as differentials of the first order so that the usual approximations employed in the theory of sound retain their validity.

The effects of heat conduction will be neglected: a simplification which may only be questionable when it is applied to low frequencies. The low frequency limit of this approximation is the characteristic frequency associated with the cooling of the heated column by conduction. This was calculated in Section 4.

The propagation of sound waves is described by a solution of the wave equation

$$\frac{\partial^2\phi}{\partial t^2} - c^2\nabla^2\phi = 0 \tag{6.1}$$

which is satisfied by the velocity potential ϕ as well as by the pressure p and the condensation s in the gas. The differential equation (6.1) is a homogeneous one, the solution $\phi(x,y,z;t)$ is determined if one knows the value of ϕ and $\partial\phi/\partial t$ at the time $t = 0$.

The physical problem to be solved here must be governed by the differential equation (6.1) in the region where no heating takes place. In the region traversed by the laser beam we expect to obtain an inhomogeneous differential equation whose solution is to be obtained for a given right side. Moreover the solution is to vanish for $t = 0$; it should represent an outgoing wave away from the region traversed by the laser beam and is to satisfy the usual continuity conditions.

The starting point in acoustics is the continuity equation for the fluid

$$\frac{\partial\rho}{\partial t} + \nabla\cdot\rho\bar{V} = 0 \tag{6.2}$$

and the equation of motion for the fluid particles:

$$\rho\,\frac{d\bar{V}}{dt} = -\,\nabla p. \tag{6.3}$$

Here the vector $\bar{V}$ denotes the velocity of the fluid, p its pressure, ρ its density. All these quantities are functions of position and time. It is customary to restrict the problem to small velocities and small displacements from static or average pressure and density p_o and ρ_o .

With these restrictions equations (6.2) and (6.3) are replaced by

$$\frac{\partial \rho}{\partial t} + \rho_o \nabla \cdot \overline{V} = 0 \tag{6.4}$$

and

$$\rho_o \frac{\partial \overline{V}}{\partial t} + \nabla p = 0. \tag{6.5}$$

We introduce the velocity potential ϕ defined by

$$\overline{V} = -\nabla \phi, \tag{6.6}$$

then (6.5) becomes

$$-\rho_o \frac{\partial}{\partial t} \nabla \phi + \nabla p = 0, \tag{6.7}$$

or

$$\nabla\left(\frac{\partial \phi}{\partial t} - \frac{p}{\rho_o}\right) = 0. \tag{6.8}$$

We write $p = p_o + p'$, where p_o is a constant and p' is a variable quantity small compared to p_o. Then

$$\frac{\partial \phi}{\partial t} = \frac{p'}{\rho_o} . \tag{6.9}$$

This last equation relates the pressure variation to the velocity potential.

Now the continuity equation (6.4) is rewritten by introducing the condensation s defined as follows

$$\rho = \rho_c(1 + s), \tag{6.10}$$

or

$$s = \frac{\rho - \rho_o}{\rho_o} \tag{6.11}$$

Then (6.4) becomes

$$\frac{\partial s}{\partial t} + \nabla \cdot V = 0, \tag{6.12}$$

or

$$\frac{\partial s}{\partial t} = \nabla^2 \phi. \qquad (6.13)$$

In the classical case the equation of sound propagation [Eq. (6.1)] is found by assuming that the pressure variation p' is proportional to the condensation s. The factor of proportionality is found by thermodynamic argument to be equal to γp_o. With this result one gets

$$p' = \gamma p_o s. \qquad (6.14)$$

Hence from (6.9)

$$\frac{\partial^2 \phi}{\partial t^2} = \frac{\gamma p_o}{\rho_o} \frac{\partial s}{\partial t}. \qquad (6.15)$$

Finally

$$\frac{\partial^2 \phi}{\partial t^2} = \frac{\gamma p_o}{\rho_o} \nabla^2 \phi, \qquad (6.16)$$

which is identical with equation (6.1). Its derivation is based on the assumption that no thermal exchange takes place.

When the gas is locally heated equation (6.14) is no longer valid. We must calculate the change in pressure when both the volume is changed and a certain amount of heat is added. If the heat is added reversibly, we write $dQ = TdS$. Then

$$dp = \left(\frac{\partial p}{\partial V}\right)_S dV + \left(\frac{\partial p}{\partial S}\right)_V dS. \qquad (6.17)$$

Equation (6.14) is a special case of (6.17) for $dS = 0$.

In the general case the coefficients of the differentials in (6.17) are evaluated as follows: The entropy change of 1 mole of an ideal gas may be written in the following two forms:[9]

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9. M.W. Zemansky: Heat and Thermodynamics, McGraw-Hill, New York, 1943, Sections 13.2 and 13.3.

$$dS = C_p \frac{dT}{T} - R \frac{dp}{p}, \tag{6.18}$$

$$dS = C_v \frac{dT}{T} + \frac{p}{T} dV. \tag{6.19}$$

Upon elimination of dT from (6.18) and (6.19) we obtain

$$\frac{R}{p} dp = (\gamma - 1)dS - \gamma\frac{p}{T} dV. \tag{6.20}$$

Hence from (6.20) and the gas law pV = RT it follows that

$$dp = - \frac{\gamma p}{V} dV + \frac{(\gamma - 1)T}{V} dS, \tag{6.21}$$

and therefore

$$\left(\frac{\partial p}{\partial V}\right)_S = - \frac{\gamma p}{V}, \tag{6.22}$$

$$\left(\frac{\partial p}{\partial S}\right)_V = \frac{\gamma - 1}{V} T. \tag{6.23}$$

We wish to express the pressure change p' in terms of the condensation s and the heat added per unit volume q. These quantities are related to dV and dS as follows

$$s = \frac{d\rho}{\rho} = - \frac{dV}{V}, \quad q = \frac{TdS}{V}.$$

Thus

$$p' = \gamma p_o s + (\gamma - 1)q. \tag{6.24}$$

Equation (6.24) must be used in place of (6.14) to obtain the equations valid in the case of the heated gas column. Differentiating (6.9) we have

$$\frac{\partial^2 \phi}{\partial t^2} = \frac{1}{\rho_o} \frac{\partial p'}{\partial t} = \frac{\gamma p_o}{\rho_o} \frac{\partial s}{\partial t} + \frac{\gamma - 1}{\rho_o} \frac{\partial q}{\partial t} . \tag{6.25}$$

On combining (6.25) with the continuity equation (6.13), we get

$$\frac{\partial^2\phi}{\partial t^2} = \frac{\gamma p_o}{\rho_o}\nabla^2\phi + \frac{\gamma - 1}{\rho_o}\frac{\partial q}{\partial t}. \qquad (6.26)$$

In the absence of heat transfer by conduction $\partial q/\partial t$ is equal to q', the rate of heat generation per unit volume. When thermal conduction is taken into account then

$$\frac{\partial q}{\partial t} = q' + K\nabla^2 T, \qquad (6.27)$$

in accordance with the heat balance equations (3.1) and (3.2). Thus, when conduction is neglected, we have

$$\frac{\partial^2\phi}{\partial t^2} - c^2\nabla^2\phi = \frac{\gamma - 1}{\rho_o} q', \qquad (6.28)$$

where $c = \sqrt{\gamma p_o/\rho_o}$ is the velocity of sound. It is apparent from this equation that of the energy communicated to the gas only the fraction $\gamma - 1$ is converted into an acoustic (hydrodynamic) wave. In the case of diatomic gases, this fraction is 0.40. Given a pulsed laser of such total output that Q is the energy incident upon the test gas during a period τ, the rate of heat deposition per unit volume is

$$q' = \alpha Q/A\tau, \qquad (6.29)$$

where A is the cross sectional area of the laser beam and α the absorption coefficient. The differential equation (6.28) may then put in the form

$$\nabla^2\phi - \frac{1}{c^2}\frac{\partial^2\phi}{\partial t^2} = - g(x,y,z,t), \qquad (6.30)$$

where

$$g = \frac{\gamma - 1}{\rho_o c^2}\frac{\alpha Q}{A\tau} f(x,y,z,t). \qquad (6.31)$$

Here f denotes a function which is 1 within the laser beam during the

duration of the pulse and 0 at every other point in space or outside the time interval $0 < t < \tau$.

The solution of this differential equation depends on the initial and boundary conditions. We assume the homogeneous initial conditions $\phi(x,y,z,0) = 0$ and $\phi_t(x,y,z,0) = 0$ which correspond to no disturbance at $t = 0$. We then have a standard radiation problem solvable in terms of the retarded potentials. In the absence of boundary conditions; i.e. for a homogeneous medium of infinite extent, the solution of (6.30) for homogeneous boundary conditions is given by the formula:[10]

$$\phi(x,y,z,t) = \frac{1}{4\pi} \iiint g(\xi,\eta,\zeta,t-r/c) r^{-1}\, d\xi d\eta d\zeta, \tag{6.32}$$

where $r = \sqrt{(x-\xi)^2 + (y-\eta)^2 + (z-\zeta)^2}$. The function g is a constant times a step function. We write Eq. (6.31) in the form

$$g = \frac{1}{\tau_1} f(x,y,z,t), \tag{6.33}$$

where

$$\tau_1 = \frac{A\rho_0 c^2 \tau}{(\gamma-1)\alpha Q} \tag{6.34}$$

is a constant that has the dimension of time. Then

$$\phi(x,y,z,t) = \frac{1}{4\pi\tau_1} \iiint \frac{f(\xi,\eta,\zeta,t-r/c)}{r}\, d\xi d\eta d\zeta. \tag{6.35}$$

The integration is to be extended over the volume of space swept by the laser beam. Its result depends only on the beam cross section and the length of the pulse.

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10. A.N. Tikhonov and A.A. Samarskii, Equations of Mathematical Physics, MacMillan, New York, 1963, pp. 461-465.

To evaluate the integral in (6.35) we place the z-axis along the direction of the propagation of the laser beam and we introduce cylindrical coordinates R, ϕ, and z. Assuming circular symmetry, we obtain readily

$$\phi(R,t) = \frac{A}{4\pi\tau_1} \int_G \frac{d\zeta}{\sqrt{R^2+(z-\zeta)^2}}, \tag{6.36}$$

where the region of integration is confined to such values of ζ for which $0 < t - r/c \leq \tau$, or equivalently

$$t - \tau < r/c \leq t. \tag{6.37}$$

Equation (6.36) is an approximation valid when the distance R from the center of the laser beam is much larger than the diameter of the laser beam, i.e. when one may replace $(x-\xi)^2 + (y-\eta)^2$ by $x^2 + y^2 = R^2$ for every point (ξ,η,ζ) within the laser beam. We introduce the new variable $\zeta' = z-\zeta$, then equation (6.36) becomes

$$\phi(R,t) = \frac{A}{4\pi\tau_1} \int_{G'} \frac{d\zeta'}{\sqrt{R^2+\zeta'^2}} \tag{6.38}$$

where the range of integration over ζ' is adjusted to be consistent with (6.37). This consistency means that

$$c^2(t-\tau)^2 - R^2 < \zeta'^2 \leq c^2t^2 - R^2. \tag{6.39}$$

The integration in (6.38) is symmetric about $\zeta' = 0$, therefore we write

$$\phi(R,t) = \frac{A}{2\pi\tau_1} \int_{\mu_1}^{\mu_2} \frac{d\zeta'}{\sqrt{R^2+\zeta'^2}} \tag{6.40}$$

where μ_1 and μ_2 are non-negative limits consistent with (6.39). We can now distinguish three regions:

a) When $R > ct$, the disturbance has not reached the observer,

$\mu_2 = 0$ and $\phi = 0$.

b) When $ct > R > c(t-\tau)$, the head of the disturbance has reached the observer, but its tail has not. In this case $\mu_1 = 0$, $\mu_2 \neq 0$ and

$$\phi = \frac{A}{2\pi\tau_1} \log \frac{ct+\sqrt{c^2t^2-R^2}}{R} . \tag{6.41}$$

c) When $R < c(t-\tau)$, the tail of the disturbance has begun to reach the observer. In this case $\mu \neq 0$ and $\mu \neq 0$ and

$$\phi = \frac{A}{2\pi\tau_1} \{\log (ct+\sqrt{c^2t^2-R^2}) - \log [c(t-\tau) + \sqrt{c^2(t-\tau)^2-R^2}] \tag{6.42}$$

It is convenient to introduce the variable $R' = R/c$ and to write ϕ in the following forms:

$$\phi(cR',t) = \frac{A}{2\pi\tau_1} \log \frac{t+\sqrt{t^2-R'^2}}{R'} , \tag{6.43}$$

for $t-\tau < R' < t$, and

$$\phi(cR',t) = \frac{A}{2\pi\tau_1} \{\log (t+\sqrt{t^2-R'^2}) - \log [t-\tau+\sqrt{(t-\tau)^2-R'^2}]\} \tag{6.44}$$

for $R' < t - \tau$. The graph of ϕ as a function of the variable t is sketched in Figure 1 for two different values of R'. The spectral distribution of the function ϕ is clearly not the same as that of the source. The function ϕ is not a square wave. Its shape depends on the distance R'. It is expected intuitively that an observer near the z-axis would observe something more resembling a square wave than an observer located farther away in a region where the sharp corners of the excitation function are washed out. The spectral distribution is calculated as follows:

The time derivative of ϕ has a very simple form. In fact,

$$\phi' = \frac{\partial\phi}{\partial t} = 0 \text{ for } R' > t,$$

$$= \frac{A}{2\pi\tau_1}\frac{1}{\sqrt{t^2-R'^2}} \quad \text{for } t > R' > t-\tau, \tag{6.45}$$

$$= \frac{A}{2\pi\tau_1}\left[\frac{1}{\sqrt{t^2-R'^2}} - \frac{1}{2\sqrt{(t-\tau)^2-R'^2}}\right] \quad \text{for } t-\tau > R'.$$

The following equation is well known:

$$\int_0^\infty J_o(bx)\sin axdx = \begin{cases} 0 & \text{for } a < b, \\ \\ \dfrac{1}{\sqrt{a^2-b^2}} & \text{for } a > b. \end{cases} \tag{6.46}$$

Replacing b by R' and a by t we have

$$\int_0^\infty J_o(R'x)\sin txdx = 0 \tag{6.47}$$

for 0 t < R'. We shall restrict our observations to the region $R' > \tau$, which is the region of the space outside of the cylinder reached by the leading edge at the disturbance when the pulse ends. We integrate (6.47) from $t = 0$ to $t = \tau < R'$ and obtain

$$\int_0^\infty \frac{J_o(R'x)}{x}(1 - \cos \tau x)\, dx = 0. \tag{6.48}$$

From (6.45) and (6.46) we find that ϕ' may be written as follows

$$\frac{\partial\phi}{\partial t} = \frac{A}{2\pi\tau_1}\left[\int_0^\infty J_o(R'x)\sin txdx - \int_0^\infty J_o(R'x)\sin(t-\tau)xdx\right]. \tag{6.49}$$

Integrate according to t from $t = 0$ to $t = s$ and note that $\phi(cR',0) = 0$.

Then

$$\phi(cR',s) = \frac{A}{2\pi\tau_1} \int_0^{\infty} J_o(R'x) \left[\frac{\cos(t-\tau)x}{x} - \frac{\cos tx}{x}\right]_o^s dx \qquad (6.50)$$

Although $J_o(R'x) \frac{\cos sx}{x}$ is not integrable from 0 to ∞, the integral in (6.50) is convergent because the expression in the brackets is

$$\frac{\cos (s-\tau)x - 1}{x} + \frac{1 - \cos \tau x}{x} + \frac{1 - \cos sx}{x} ,$$

and the product of each of these functions with $J_o(R'x)$ is integrable. In fact the middle term contributes nothing (see eq. 6.48). Then

$$\phi(cR',s) = \frac{A}{2\pi\tau_1} \int_0^{\infty} \frac{J_o(R'x)}{x} [\cos (s-\tau)x - \cos sx]\, dx. \qquad (6.51)$$

Hence, using an elementary trigonometric identity, we get

$$\phi(cR',s) = \frac{A}{\pi\tau_1} \int_0^{\infty} \frac{J_o(R'x)}{x} \sin \frac{\tau x}{x} \sin (s-\frac{\tau}{2})x\, dx. \qquad (6.52)$$

Now let $t = s - \frac{\tau}{2}$ and $x = \omega$, then

$$\phi(cR',t+\frac{\tau}{2}) = \frac{A}{\pi\tau_1} \int_0^{\infty} \frac{J_o(R'\omega)}{\omega} \sin \frac{\omega t}{2} \sin \omega t\, d\omega. \qquad (6.53)$$

We have represented $\phi(cR',t+\frac{\tau}{2})$ as a Fourier integral. The Fourier sine transform of ϕ is thus

$$\phi(\omega) = \frac{A}{\pi\tau_1} J_o(R'\omega) \frac{\sin \omega\tau/2}{\omega} . \qquad (6.54)$$

Here we define the sine transform by the relations

$$F(\omega) = \frac{2}{\pi} \int_0^{\infty} f(t) \sin \omega t\, dt, \text{ and } f(t) = \int^{\infty} F(\omega) \sin \omega t\, d\omega.$$

The spectral density of $\phi(cR',t)$ is therefore

$$\phi(\omega)^2 = J_o^2(R'\omega)\left(\frac{\sin \omega\tau/2}{\pi}\right)^2 \frac{A^2}{\pi^2 \tau_1{}^2} \tag{6.55}$$

It is shown by elementary calculations that the spectral density function of a square wave of unit amplitude and of duration τ is

$$\phi_s(\omega)^2 = \left(\frac{4}{\pi}\right)^2 \left(\frac{\sin \omega\tau/2}{\omega}\right)^2 . \tag{6.56}$$

Comparison of (6.55) and (6.56) shows that in our problem the spectrum of the square wave gets distorted by the factor $J_o(R'\omega)$. The distortion is small near the source where $J_o \sim 1$. For $R'\omega \sim 2.4$, however, $J_o = 0$. This means that at a given distance R the frequency for which R = 2.4c will be missing. Other missing frequencies correspond to other zeros of J_o. The reduction in the spectral intensity due to the $J_o(R'\omega)$ factor can be calculated from the following table which gives the reduction from the intensity of a square pulse. The entry item of the table is the product $R\nu$ in cm times cycles per second. Assumed velocity $c = 3.3 \times 10^4$ cm/sec.

νR(cm/sec)	500	1000	1500	2000	2500	3000	3500	4000	4500
$J_o^2\left(\frac{2\pi\nu R}{c}\right)$	0.996	0.980	0.960	0.929	0.891	0.846	0.794	0.741	0.680

It is clear that only for large distances or high frequencies will the Bessel factor affect significantly the spectral distribution of the energy. The main determining factor at small distances and moderate frequencies is the spectral distribution of the pulse. This distribution is characterized by the function

$$\left(\frac{\sin \frac{\omega\tau}{2}}{\frac{\omega\tau}{2}}\right)^2$$

which is well known from the theory of diffraction. It is sketched in Fig. 6.2. The largest part of the energy is contained in the spectrum region between 0 and $\omega\tau = 2\pi$. Thus, in terms of frequency, the interval $0 < \nu < 1/\tau$ contains most of the energy. Moreover, it is manifest from Fig. 6.2 that the spectral distribution is rather flat in the region in which most of the energy is contained. A pulse of this type is not suitable as a source for a signal which is to be detected by a narrow-band detector.

The question arises, what happens when a pulse of the type described is repeated periodically at equal time intervals p. In this case the Fourier integral is replaced by a Fourier series and the continuous spectral distribution shown in Fig. 6.2 is replaced by a discrete spectrum consisting of equidistant lines with the separation $\Delta\nu = 1/p$. The relation of this discrete spectrum to the continuous spectrum of the non-periodic (single pulse) case is illustrated in Fig. 6.3. Again the energy is generally broadly distributed and only for $p \sim 2\tau$ does a significant spectral concentration of energy take place.

Note: It is possible to remove the restriction that the observation should be confined to the region $R' > \tau$. This assumption was made following equation (6.47). In the general case we cannot assert that the left hand side of equation (6.48) is zero, but we can assert that it is a function of R' and τ alone. Let it be denoted by $\psi(R',\tau)$. Then we can carry out the calculations that lead to equations (6.52) and (6.53) provided we replace $\phi(cR',s)$ by $\phi(cR',s) + \psi(R',\tau)$, where the function ψ is independent of t. Consequently the spectrum of $\phi(cR',t)$ will be the same as before since the time independent term can not affect $\phi(\omega)$ for $\omega \neq 0$.

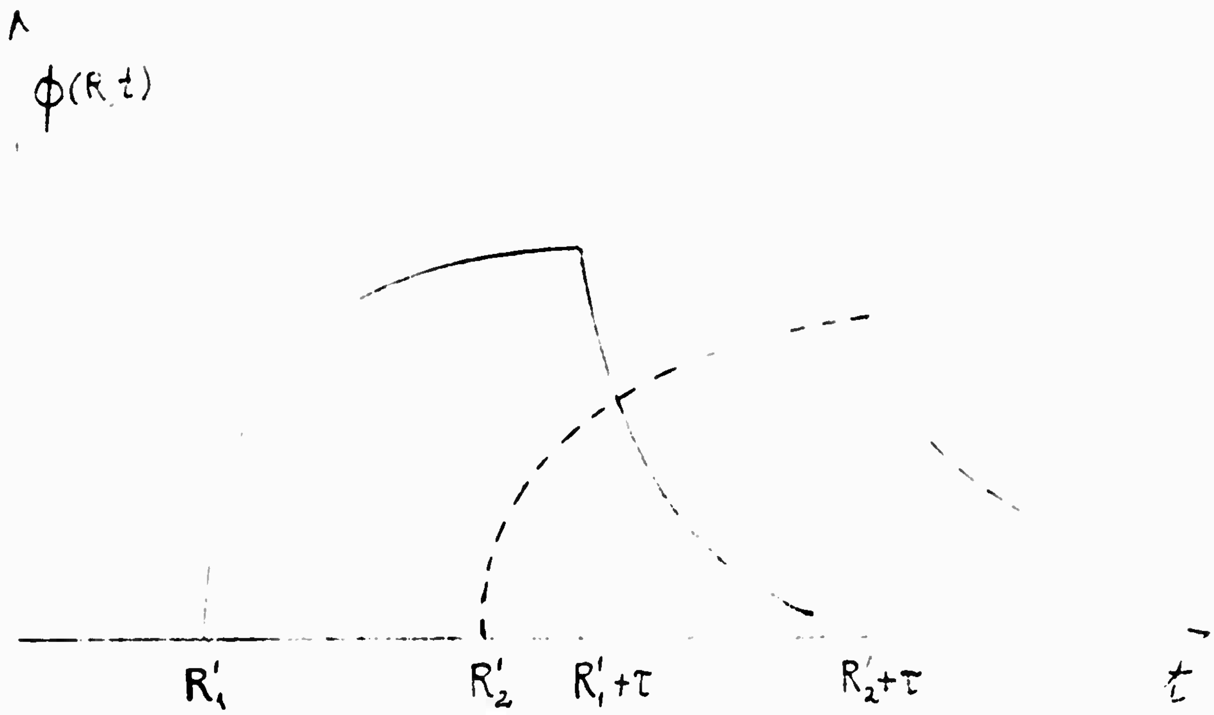

Figure 6.1. Distortion of a square pulse emanating from a line source. The function $\phi(R,t)$ defined by equation (6.40) is shown for two different values of R. ($R' = R/c$.)

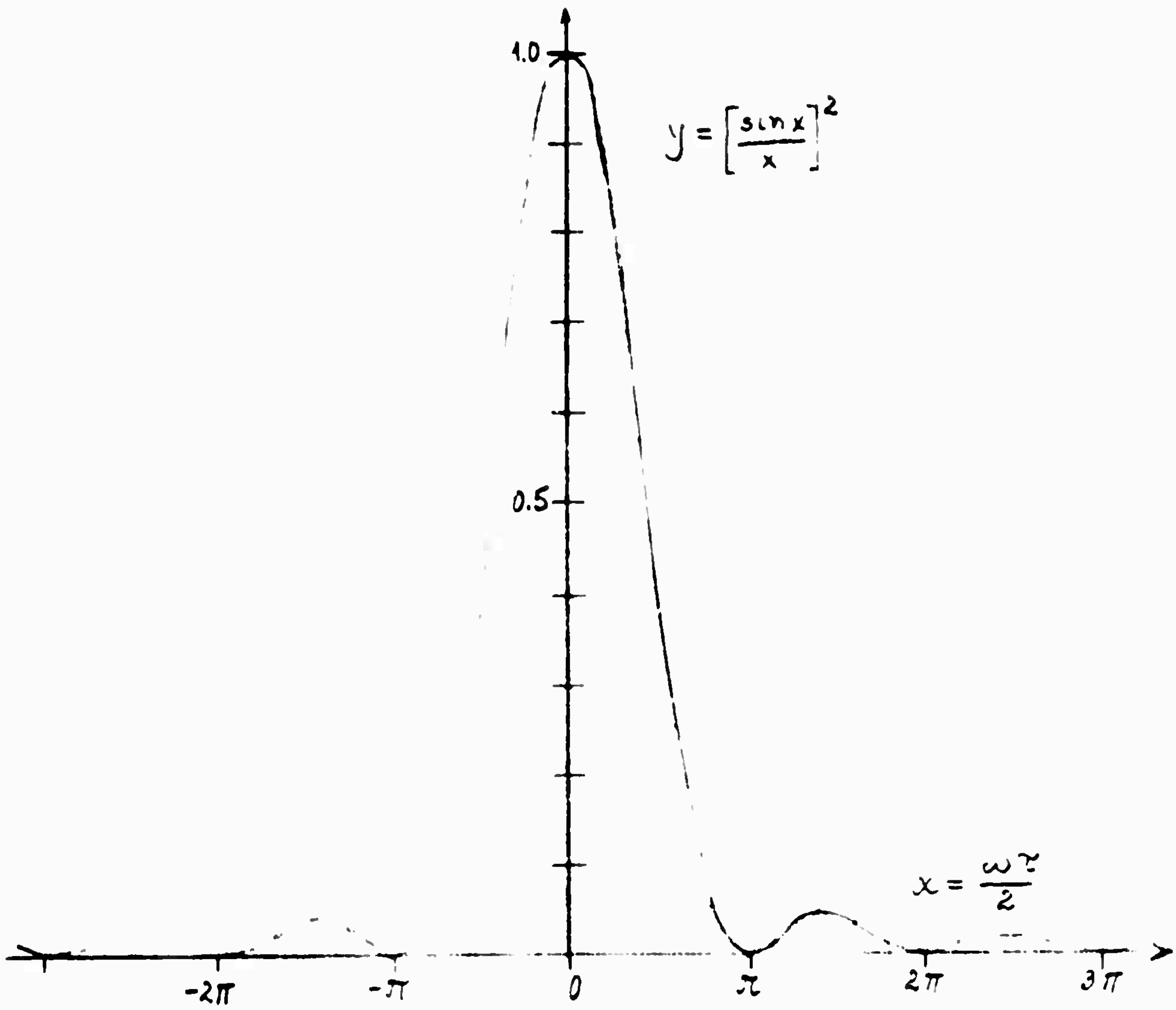

Figure 6.2. The spectral density function of a square pulse.

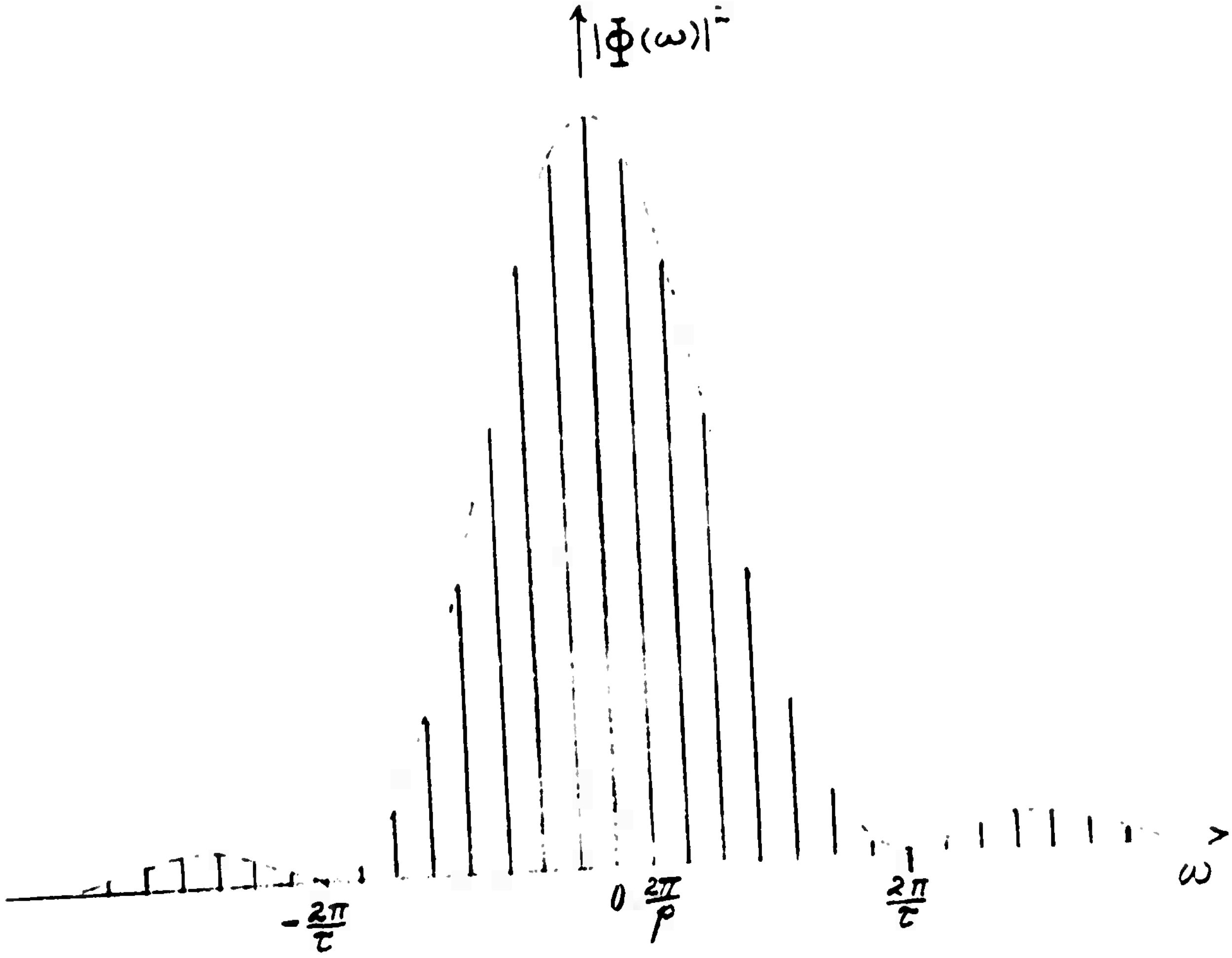

Figure 6.3. Fourier analysis of a square pulse of length τ repeated periodically at intervals p.

7. The Acoustic Problem in a Confined Region.

In the preceeding section we analyzed the generation of a sound pulse from the thermal energy deposited in a gas by laser pulses. No boundary conditions were imposed, thus implying that the gas was not confined. If the gas were not confined, or if the confining walls were far removed from the path of the beam, acoustic energy would radiate in all directions. The intensity of the acoustic signal would then decrease as the reciprocal of the distance from the center line of the generating beam. The detection or measurement of such a signal depends to a large extent on the experimental arrangement used to concentrate a significant fraction of the signal generated on a suitable detector capable of converting the incident acoustic signal into an electric signal. It would require a rather sophisticated, well-designed system to utilize as much as 20% of the generated acoustic energy. Even if much of the energy radiated out is concentrated on the detector, by means of reflectors, for example, a serious degradation of the detector system will result from the fact that the generated signal has a rather broad spectral distribution.

Now we shall examine the generation of sound in a confined system, a cylinder concentric with the laser beam. It is expected that in such a system the efficiency of the utilization of the sound generated will be higher, and one might hope to make use of the resonances of the confining

tube to enhance detection. The following questions are worth examining:

1. How does the confining tube affect the generation and propagation of the sound wave?
2. Is it possible to enhance the measurement of the sound wave generated by proper choice of the geometry of the confining vessel?

The starting point of the analysis is the differential equation (6.30)

$$\nabla^2\phi - \frac{1}{c^2}\frac{\partial^2\phi}{\partial t^2} = - g(x,y,z,t) \tag{7.1}$$

with the initial conditions

$$\phi(x,y,z,0) = 0; \ \phi_t(x,y,z,0) = 0. \tag{7.2}$$

The meaning of the variables was explained in section 6, where it was also noted that g is zero except in the part of the space swept out by the laser beam. In this cylindrical region, g has a constant value during the time interval $0 < t \leq \tau$; it is 0 when $t > \tau$. We shall simplify the computations by normalizing g so that is has the values 0 and 1. Since g has the dimension of reciprocal time, it is more correct to say that we set the peak value of g at 1 sec^{-1}.

The presence of the walls of the tube requires the introduction of a boundary condition which expresses the fact the normal component of the fluid velocity at the walls is zero. This means

$$\overline{n}\cdot\nabla\phi = 0. \tag{7.3}$$

at all boundaries. We shall seek a solution of (7.1) in the region $x^2 + y^2 \leq b^2$ for $t \geq 0$. A useful technique for the solution of such a problem is the eigenfunction expansion. This technique relies on the determination first of the simple harmonic solutions of the homogeneous equation

obtained from (7.1) with $g \equiv 0$. It is well known that, if we set

$$\phi = e^{-i\omega t}\, \psi(x,y,z), \tag{7.4}$$

then ψ satisfies the homogeneous Helmholtz equation

$$\nabla^2\psi + k^2\psi = 0 \tag{7.5}$$

which is obtained from (7.1) by setting $g = 0$, substituting (7.4) and $k = \omega/c$. It is also known that, given a finite region $\mathcal{R}$ and given homogeneous boundary conditions, non-vanishing solutions of (7.4) are possible only for a discrete set of values of k, called the eigenvalues of (7.4) and that solutions corresponding to different eigenvalues are orthogonal to each other in the sense

$$\int \psi_n\, \psi_m^{*}\, dv = 0, \tag{7.6}$$

whenever $k_n \neq k_m$. Following standard practices in mathematical physics, we obtain an ascending, infinite sequence of eigenvalues

$$k_1 \leq k_2 \leq k_3 \leq \cdots, \tag{7.7}$$

and a corresponding sequence of orthogonal and dimensionless eigenfunctions

$$\psi_1,\ \psi_2,\ \cdots$$

so normalized that they satisfy the relations

$$\int \psi_i\, \psi_j^{*}\, dv = \delta_{ij}\, V, \tag{7.8}$$

where $dv = dx\, dy\, dz$ and integration is over the entire volume V of region $\mathcal{R}$. It is known that the sequence of such functions is complete for equations of the type we are considering and that a well-behaved function of x, y, z is expandable in the region $\mathcal{R}$ in terms of these eigenfunctions.

Convergence in the mean is assured by the standard theorems of functional analysis.

With these preliminaries we can reduce the solution of our original problem to the calculation of the excitation of an infinite number of oscillators by a given exciting force. We write

$$\phi = \sum \mu_n(t)\psi_n(x,y,z), \tag{7.9}$$

$$g = \sum g_n(t)\psi_n(x,y,z). \tag{7.10}$$

The coefficients $\mu_n(t)$ and $g_n(t)$ are calculated as follows:

$$V\ \mu_n = \int \phi\psi_n^* \, dv, \tag{7.11}$$

$$V\ g_n = \int g\psi_n^* \, dv. \tag{7.12}$$

The differential equation (7.1) then requires that

$$\mu_n'' + \omega_n^2\ \mu_n = g_n \tag{7.13}$$

for $n = 1,2,3, \cdots$. Since the g_n's are calculable from (7.12), the solution of the complete problem now reduces to the determination of the excitations of the harmonic oscillators whose equations are given above. The excitations depend on the characteristic frequencies of these oscillators, ω_n, and on the functions $g_n(t)$.

The total energy fed into the n-th oscillator is proportional to

$$\int_0^\infty |g_n(t)|^2 dt$$

and to another factor which represents the response of the n-th oscillator to a normalized exciting force. This response factor depends on ω_n and on

the time dependence of the exciting force g_n. (It is to be noted that g_n has the same dimension as g, which is not that of force. At the same time, the dimension of μ_n is that of ϕ.)

In principle the calculations require the solution of infinitely many differential equations of the form (7.13) for very general functions $g_n(t)$ on their right side. Practically the situation is much simplified because the physically relevant part of the solution may be obtained by solving only one or a few equations of the type described. There are two main reasons for this simplification. First, in a situation with some degree of symmetry many of the functions $g_n(t)$ are identically zero, because the spatial part of the excitation function is orthogonal to many ψ_m's. Second, the sequence k_1, k_2, $\cdots$ increases rather rapidly and consequently only a few of the harmonic oscillators which are described by equation (7.13) have resonant frequencies in the region in which f has a significant harmonic component.

We shall return to the theory after exhibiting some of the well-known eigenvalues and eigenfunctions appropriate for sound propagation in a long circular cylinder.

The eigenfunctions appropriate for a right circular cylinder of length L and radius b are the non-vanishing solutions of equation (7.5) which satisfy the boundary conditions

$$\frac{\partial\psi}{\partial r} = 0 \text{ for } r = b, \text{ and } \frac{\partial\psi}{\partial z} = 0 \text{ for } z = \pm L/2.$$

After separating equation (7.5) in cylindrical coordinates, one obtains

solutions of the form

$$\psi_s = e^{in\theta} J_n(\sqrt{k^2-h^2}r) \cos hz \tag{7.14}$$

and

$$\psi_a = e^{in\theta} J_n(\sqrt{k^2-h^2}r) \sin hz, \tag{7.15}$$

where the subscript s denotes a function symmetric for the reversal of the z-axis, while subscript a denotes an antisymmetric function. J_n is the Bessel function of order n, where n must be an integer to insure uniqueness of the solution. These functions are not yet normalized. We note that if the cylinder contained one or more septum; i.e. a boundary plane containing the cylinder axis, then n need not be an integer. We shall avoid this situation.

The possible values of h are determined by the boundary conditions at the cylinder ends. These require that, for symmetric functions sin hL/2 = 0; i.e.

$$h_\ell = 2\ell\pi/L;\ \ell = 0,1,2,\cdots. \tag{7.16}$$

For antisymmetric functions cos hL/2 = 0; i.e.

$$h_\ell = (2\ell + 1)\pi/L;\ \ell = 0,1,2,\cdots. \tag{7.17}$$

Boundary conditions on the cylinder surface r = b require that

$$J_n'(\sqrt{k^2-h^2}b) = 0. \tag{7.18}$$

Therefore k can only have such values for which the number $\chi = \sqrt{k^2-h^2}b$ is a solution of the equation $J_n'(\chi) = 0$. For each index n, the derivative of the n-th order Bessel function has an infinite number of discrete positive roots. We write these in ascending order as χ_{nm}; $m = 1,2,\cdots$. The eigenvalues of the differential equation (7.5) consistent with the

boundary conditions described above are calculated from the equations

$$(k^2_{nm\ell} - h^2_\ell)b^2 = \chi^2_{nm};\ n = 0,1,\cdots;\ m = 1,2,\cdots, \tag{7.19}$$

where h_ℓ are the numbers determined from (7.16) and (7.17). The symmetric eigenfunctions are

$$\psi_{snm\ell} = e^{in\theta}\ J_n(\chi_{nm}r/b)\cos h_\ell z, \tag{7.20}$$

while the antisymmetric eigenfunctions are

$$\psi_{anm\ell} = e^{in\theta}\ J_n(\chi_{nm}r/b)\sin h_\ell z. \tag{7.21}$$

Of the triple infinity of eigenfunctions (or oscillation modes) given in (7.20) and (7.21), certain types are of particular interest to us. Only $\ell = 0$ produces z-independent eigenfunctions. These are called transverse modes. Their equations are

$$\psi_{snm} = e^{in\theta}\ J_n(\chi_{nm}r/b). \tag{7.22}$$

Modes which have rotational symmetry about the z-axis are all of the form

$$\psi_{som\ell} = J_o(\chi_{om}r/b)\cos\frac{2\pi\ell}{L}z \tag{7.23}$$

and

$$\psi_{aom\ell} = J_o(\chi_{om}r/b)\sin\frac{(2\ell+1)\pi}{2L}z \tag{7.24}$$

The first few roots of $J_o'(r) = 0$ are 3.832, 7.016, 10.173, 13.324. Those of $J_1'(r) = 0$ are 1.841, 5.330, etc.[11]

Let us now calculate the excitation of a cylindrical resonator with a force function that depends only on r and t; i.e. which is independent

11. See Ref. 8.

of θ and z. In this case every integral of the type

$$\int_0^b \int_0^{2\pi} g(r,t)\ J_n(\chi_{nm} r/b) e^{in\theta}\ r dr d\theta$$

vanishes for $n \neq 0$, leaving only the eigenfunctions with $n = 0$ to interact with g. Similarly the integrals over z involving the product of g with $\sin h_\ell z$ and $\cos h_\ell z$ will vanish except for $\cos 0z$. Thus, when evaluating the expansion of $g(r,t)$ in terms of the eigenfunctions of the problem, only the symmetric eigenfunctions with $n = 0$ and $\ell = 0$ need to be considered. The relevant eigenfunctions are $J_o(\chi_{om} r/b)$. Their normalization factor is calculated from the standard formula[12]

$$\int_0^b [J_o(\chi_{om} r/b)]^2 r dr = \frac{1}{2} b^2 [J_o(\chi_{om})]^2, \qquad (7.25)$$

valid for every χ_{om} for which $J_o'(\chi_{om}) = 0$. The normalized functions are

$$\psi_m = J_o(\chi_{om} r/b)/J_o(\chi_{om}). \qquad (7.26)$$

When the excitation is of unit magnitude in a small cylinder of radius a centered around the polar axis and lasts from $t = 0$ to $t = \tau$, we have from (7.12) and (7.26)

$$Vg_m(t) = \frac{2\pi L}{J_o(\chi_{om})} \int_0^a J\ (\chi_{om} r/b) r dr f(t), \qquad (7.27)$$

where $f(1) = 1\ \mathrm{sec}^{-1}$ for $0 < t < \tau$, and 0 for $t > \tau$. For small values of a we may replace $J_o(\chi_{om} r/b)$ by 1 and obtain the approximation

$$g_m(t) = \frac{v}{V} \frac{f(t)}{J_o(\chi_{om})}, \qquad (7.28)$$

where $v = a^2 \pi L$ is the volume swept out by the laser beam.

12. Ref. 8, p.91.

The next problem is to deal with a set of harmonic oscillators with pulse excitation. Before losses in the oscillators are considered, their equations have the form

$$\mu_m'' + \omega^2\mu_m = g_m, \tag{7.29}$$

where g_m is given in (7.28). The characteristic frequencies are

$$\omega_m = \chi_{om}c/b. \tag{7.30}$$

Taking $c = 3.3 \times 10^4$ cm/sec, we get for the lowest symmetric transverse frequency (corresponding to $\chi_{o1} = 3.832$)

$$\nu_1 = \frac{2.0\text{x}10^4}{b},$$

and for the next lowest

$$\nu_2 = \frac{3.66\text{x}10^4}{b},$$

where b is in cm, ν in sec^{-1}. Thus the lowest such frequency for a tube 10 cm in diameter is 4000 sec^{-1}, the next 7330 sec^{-1}. A small tube of 1 cm diameter would require acoustic frequencies which are prohibitively high.

Generally the oscillators will have a certain loss rate associated with them. Losses are not only unavoidable but necessary when the resonator is a part of a system used for measurement, because measurement consists of converting a part of the acoustic disturbance into an electric signal. Thus a resonator will have an output which constitutes the measured signal, or else the energy is communicated to another frequency-sensitive device, the detector, which converts it to an electric signal.

In any case, we are interested in devices which subject the disturbance to a harmonic analysis. The noise discriminating property of the detecting system improves as its bandwidth is restricted. Unfortunately, as we shall see, narrowing the bandwidth drastically reduces the signal.

For the sake of simplicity, let us assume that the detector is the cavity itself, that is, we do away mentally with a second tuned device to which the cavity may be coupled. If there is such a separate device, the detected signal will be less because some of the energy will not be transferred and new sources of noise will be introduced. We thus have a sequence of oscillators with an output and we concentrate on the one with the lowest frequency. When the effects of oscillator loss (energy conversion) are incorporated in the equations of the harmonic oscillators, we obtain for the oscillator of the lowest frequency[13]

$$\mu'' + \Delta\mu' + \omega_1^2\mu = g_1(t). \qquad (7.31)$$

The quantity Δ has the dimension of reciprocal time. Let W denote the energy stored in the free running harmonic oscillator. It is known that

$$\Delta = \frac{dW}{dt} W^{-1} \qquad (7.32)$$

One can readily show the following: When g is a sinusoidal function of time, the excitation of the oscillator is significant only if the frequency of the exciting force is nearly equal to that of the free-running oscillator. Then by means of Fourier analysis one concludes that in the case of a general time-dependent force, the excitation of the oscillator

13. P.M. Morse, Vibration and Sound, McGraw-Hill, New York, 1948, Ch. II.

depends largely on the harmonic component of the exciting force which lies near ν_1. After a short calculation one finds that the average energy of the oscillator is approximately

$$W \approx \frac{|G(\nu_1)|^2}{2\Delta} \tag{7.33}$$

where $G(\nu)$ is the Fourier transform of the exciting function $g(t)$ and $\nu_1 = \omega_1/2\pi$. The power output of such an oscillator is

$$\frac{dW}{dt} = \Delta W \sim \frac{1}{2}|G(\nu_1)|^2 \tag{7.34}$$

Only part of this is useful power output in the detector, but this brings in only a factor of about one-half, which is not significant. The important matter is that the rate at which power is absorbed by the oscillators is proportional to the Fourier transform of the excitation function at the resonant frequencies of the oscillators.

The effect of the confining tube is that the acoustic signal generated within it is resolved into its spectral components according to the resonant frequencies of the cavity. As we have seen in section 6, the power spectrum of a single pulse of duration τ is rather broad, most of the energy is in the spectral interval 0 to $1/\tau$. Periodic repetition of the pulse produces a spectral distribution of the acoustic energy among the harmonics of the repetition frequency in the manner illustrated in Fig. 6.3.

In order to obtain good utilization of the acoustic energy generated, the experiment should be so designed that a large fraction of the acoustic energy generated by the source is concentrated in the frequency region capable of exciting a single selected cavity resonance because the detector

will only be coupled to one resonance. Such concentration of the acoustic energy is not easily achieved. With a single pulse it is just about hopeless to obtain success in this manner. With pulses repeated at the rate of a resonant frequency of the cavity, the situation is better, but reasonable cavity dimensions require rather high pulse repetition frequencies. We calculated that the lowest resonance of a cylindrical cavity of 10 cm diameter is at 4000 Hz, therefore we would have to operate with the laser pulses shorter than 0.1 msec repeated 4000 times per second. The success of the scheme of this type depends not only on the construction of a powerful laser with appropriate pulsing mechanism but also on the construction of a highly sensitive acoustic detector coupled with the proper mode of oscillation of a cylinder.

8. Interferometer Method.

It is plausible to contemplate a two laser experiment in which one high intensity laser pulse heats the air and the second probe laser beam travels the same path which is a branch of an interferometer.[14] Then the heating effect produced by the intense laser pulse would cause a shift in the interference pattern of the probe beam due to a change in the optical path length of one of the branches of the interferometer. See Figure 8.1. The absorption chamber may be evacuated to determine the effect of the intense laser pulse on the end windows; or, if measurements are to be made in the atmosphere, the end windows may be removed.

If Q is the energy transmitted in the intense pulse of cross sectional

14. Longaker and Litvak: Refractive Index Changes in Absorbing Media by a Pulsed Laser Beam. Bull. Am. Phys. Soc. 11, No. 1, 129, 1966.

area A, then the energy balance over a unit length of the gas traversed by the intense pulse gives:

$$Q\alpha = A\rho J'c_p\delta T,$$

where c_p is the specific heat of the gas at constant pressure

$J' = 4.19$ joules/cal.

δT is the increase in temperature of the gas.

Hence

$$\delta T = \frac{Q}{A}\,\frac{\alpha}{J'\rho c_p}\ .$$

The temperature increase δT produces a density change $\delta\rho$. In the case of an ideal gas, where the temperature change is assumed to take place quickly enough to occur at constant pressure,

$$\frac{\delta T}{T} + \frac{\delta\rho}{\rho} = 0.$$

The change of density causes a change in the refractive index according to the relation,

$$\frac{n-1}{n_o-1} = \frac{\rho}{\rho_o}\ ,$$

where n is the refractive index of the gas at the frequency of the probe laser beam.

From these relations it follows that

$$\delta n = (n-1)\,\frac{\delta\rho}{\rho} = -\ (n-1)\,\frac{\delta T}{T}\ ,$$

or

$$\delta n = -\ \frac{(n-1)}{T}\ \frac{Q\alpha}{AJ'\rho c_p}\ .$$

Now if L is the length of the absorption chamber, the change in optical path length for the probe, or monitor, laser beam is $L\ \delta n$, which will be a certain fraction, F, of its wave length. Hence,

$$L\ \delta n = F\lambda,$$

and one has

$$F = \frac{L}{\lambda}(n-1)\alpha \frac{Q}{AJ'\rho c_p T} .$$

The minimum value of α which can be detected depends upon how small a value of F one can measure for a given value of the other physical quantities in the experiment.

For example, assuming $Q/A = 20$ joules/cm^2, $L = 100$ cm, $\lambda = 5 \times 10^{-5}$ cm, $(n-1) = 3 \times 10^{-4}$, $T = 300°K$, $\rho = 1.3 \times 10^{-3}$ gm/cm^2, $c_p = 0.24$ cal/gm °C, one has $F \approx 3 \times 10^4\ \alpha$.

If one can measure a path length change, or phase shift, of a thousandth of a wave length ($F = 0.001$), one can then detect α of 3×10^{-8} cm^{-1}. One can increase the energy and path length to perhaps detect an α of an order of magnitude less. But a phase change of a thousandth of a wave length is difficult to measure by methods usually associated with interferometry.

In order to measure extremely small phase shifts, of the order of $\lambda/1000$, or possibly smaller, the second branch of the interferometer probe laser beam is also varied in optical path length, but at a frequency of the order of 10^5 Hz.[15] This may be accomplished by the insertion of a crystalline Kerr Cell in the optical path, or by making one mirror a Piezo-electric crystal, either one driven by a 10^5 Hz. oscillator. The interference pattern

15. P.H. Lee and Skolnick: "A Phase Comparison Optical Discriminator", IEEE J. Quantum Electronics, QE-2, No. 12, p. 784-785, 1966.

is detected by a photomultiplier tube whose output will be the 10^5 Hz. signal caused by the driven crystal. This is fed into a synchronous detector, together with a signal directly from the 10^5 Hz. oscillator. Now a servo-mechanism is used to apply a variable high D-C voltage to the crystal in order to further vary the optical path of the second branch in the same amount that the intense laser pulse varies the optical path of the first branch, thus keeping the time average optical path of the second branch equal to the optical path of the first. See Figure 8.2. The interference pattern is then kept in phase, and the D-C servo-voltage required to do this is proportional to the phase shift that would otherwise occur. Hence the small phase shift is converted into a measurable voltage.

The limitations here are determined by the constancy in optical path one can achieve by carefully constructing the interferometer and by the homogeneity of the wave fronts in the first branch through which the intense laser is pulsed. A detectivity of α down to the order of 10^{-9}/cm should be achievable under favorable conditions. This appears as the most promising alternate to the spectrophone, especially for intense laser pulses at frequencies where scattering is appreciable. Smaller values of α might be detectable by measuring effects over long ranges, as might be possible in a mile long tube.

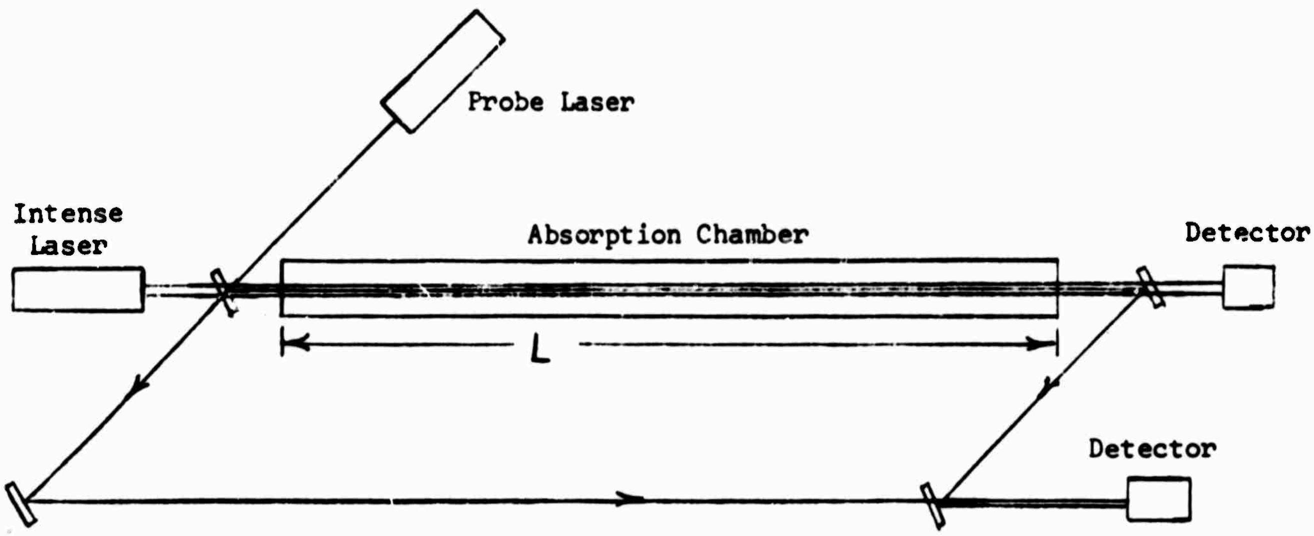

Figure 8.1. Interferometer

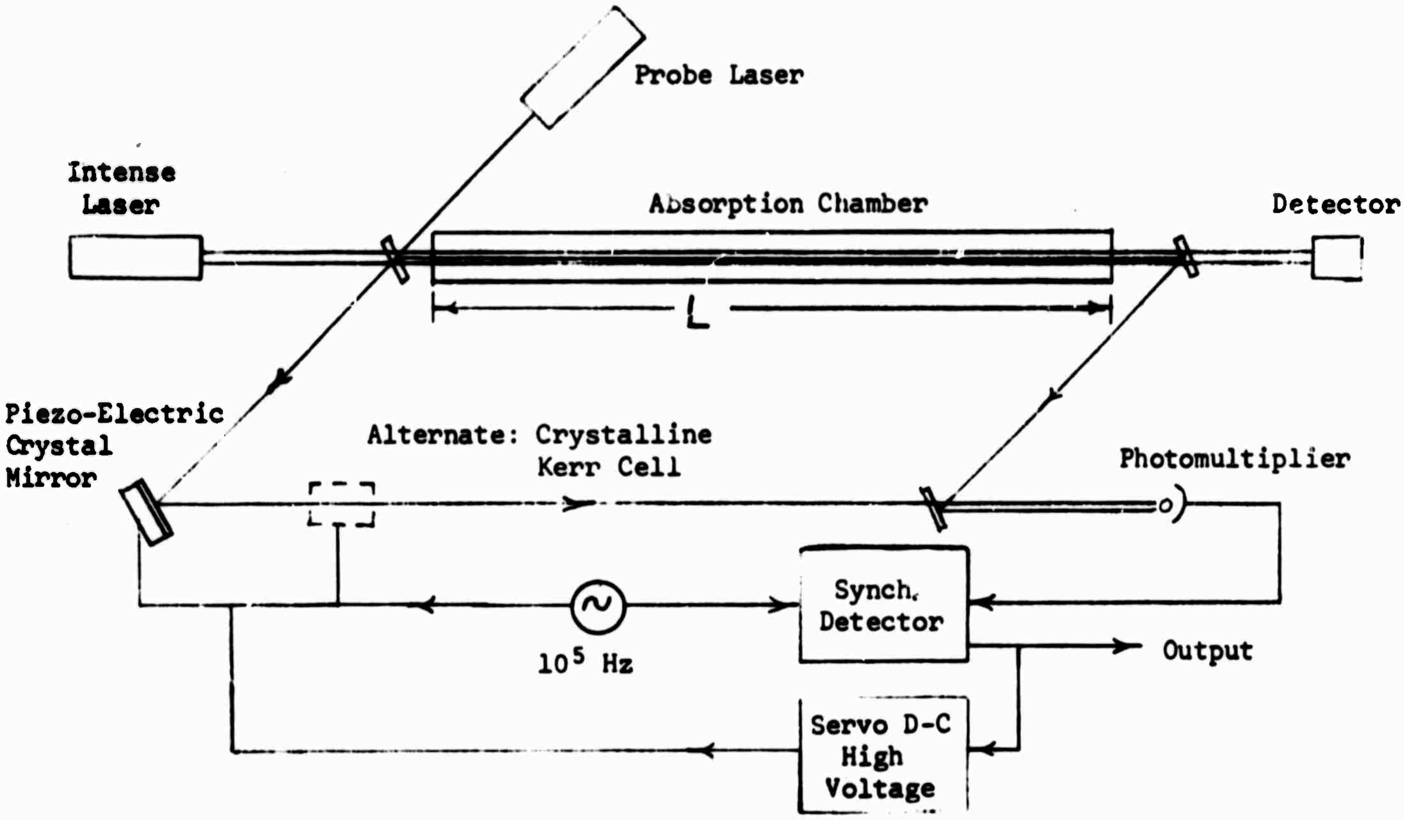

Figure 8.2. Interferometer With Phase Comparison Optics[15]

9. The Absorptivity Spectrophone and Measurement of α.

The absorptivity spectrophone, a practical, direct method for precise measurement of weak optical absorptivity in gases, especially for wave lengths at which scattering is not significant, has been designed by Edwin L. Kerr and John G. Atwood.[16] They used the spectrophone to measure the absorption coefficient of the 9.6 micron line of a CW CO_2 laser in mixtures of CO_2 - N_2. At 300 ppm CO_2 in N_2, the standard concentration of CO_2 in air at sea level, the absorptivity reported was 5.8×10^{-7} cm^{-1}.(16) The same equipment, the theory of which has been discussed in Section 3, was used by us in an attempt to measure the absorption coefficient of the 10.6 micron line of a CW CO_2 laser in the actual atmosphere. At $24° \pm 1°$ C with a relative humidity of $(70 \pm 5)\%$ and an unknown amount of air pollutants present in the air in the Western San Fernando Valley, the measured value of the atmospheric absorption coefficient is $(1.0 \pm 0.2) \times 10^{-6}$ cm^{-1}. Time and experimental difficulties did not permit the accomplishment of a more ambitious goal, that of measuring α in controlled, known mixtures of N_2 - O_2 - CO_2 - H_2O while simultaneously monitoring the spectral distribution of the laser beam.

The spectrophone measures absorptivity by sensing the thermal expansion of a confined sample of gas. A complete description of the device is contained in the Perkin-Elmer Report(16). Only a brief description is contained here. The laser was made in house, except for the pyrex tube which was loaned to us by a prominent manufacturer of same. The length is 0.7 meters with an inside diameter of 0.5 inches. Using one plane and one

16. Earl L. Sloan and Edwin L. Kerr, Perkin-Elmer Corporation Report No. 8884, Absorption of Light, 31 July 1967.

spherical mirror, it supplied an output of just under one watt during the experiments while excited by a D-C voltage of 12,000 and containing the following (closed) gas mixture:

1.0 Torr CO_2, 2.5 Torr N_2, 11.0 Torr He, 0.2 Torr H_2O.

The spectral distribution is dependent upon many factors, including proper alignment of the end mirrors. Spectral measurements were not made simultaneously with atmospheric absorption. Hence all that can be said is that measurements made at different times, when the laser is tuned for maximum power, as during the experiments, indicate that the laser power is almost entirely at 10.6 microns. Laser power was measured with a Korad Model K-PM Power Meter.

The absorptivity measurements were made by detecting the pressure rise in a gas sample chamber while the CW CO_2 laser beam is passed through. The sample chamber and pressure transducer were obtained from the Perkin-Elmer group, where they were used for similar measurements. The sample chamber is aluminum, 2 cm I.D., 10 cm O.D., 20 cm path length, 4.2 kg mass, and has 6mm KBr windows at Brewster's angle. The chamber is massive to insure a uniform constant temperature. Thermal and acoustic shielding is provided by an outer cylindrical aluminum tank which is sealed. The laser beam power was monitored by a Korad K-PM power meter. The pressure rise was measured by an MKS Baratron pressure transducer with a minimum resolvable differential of 10^{-5} mm Hg. or 10^{-8} atm. A steady state temperature distribution is reached in a few tenths of a second after the shutter opens to allow the beam to shine through the chamber.

As derived in Section 3, with the simplifying assumption of a perfectly

defined cylindrical beam, the pressure rise is:

$$dP = \frac{P_o \alpha J W}{T_o 4 \pi K}\left(1 - \frac{a^2}{2b^2}\right) \tag{3.10}$$

where P_o = initial pressure

T_o = initial temperature

α = linear absorption coefficient

W = beam power

K = thermal conductivity of gas in sample chamber

J = 0.239 cal/joule

a = beam radius

b = sample chamber radius.

In actual practice, the power density of the laser beam is approximately a Gaussian distribution, with the intensity given by:

$$I(\rho) = \frac{W}{4\pi w^2} e^{-\rho^2/w^2}$$

where w = half beam width at e^{-1} points, ρ = distance from axis. With this refinement, the quantity in parenthesis $(1 - \frac{a^2}{2b^2})$ is replaced by [16]

$$\left[1 - \frac{w^2}{b^2} + \left(\frac{w^2}{b^2} - \frac{b^2}{2w^2}\right) e^{-b^2/w^2}\right] \approx \left(1 - \frac{w^2}{b^2}\right) \text{ for } w < b.$$

The reproducibility of measurement of the pressure rise, dP, was of the order of 10%, hence a precise measurement of w is unnecessary, as $(1 - \frac{w^2}{b^2}) \approx 0.98 \pm 0.01$ in our experiment.

The absorption chamber was baked out at temperatures in excess of 100°C purged with dry nitrogen several times to attempt to rid the KBr windows of contaminants, but enough remained to give an equivalent absorptivity of

10^{-7} cm^{-1} with dry N_2 at atmospheric pressure in the cell. The N_2 was evacuated and air was admitted to the cell, and absorptivity measurements were made on several days -- all of which were midly smoggy. With the consistent weather in the Western San Fernando Valley, the climate conditions were similar on all days. Temperature (24 $\pm$ 1)°C. Pressure 740 $\pm$ 5 mm Hg. Relative humidity (70 $\pm$ 5)%. Very mild smog conditions. Under these conditions the measured value of $\alpha = (1.0 \pm 0.2) \times 10^{-6}$ cm^{-1}.

10. Conclusions.

The following conclusions were reached concerning the measurement of the absorption of radiation in air for wave length regions where $\alpha < 10^{-8}$ cm^{-1}:

1. It is probably possible to make absorption measurements in air using a steady laser beam and noting the static pressure increase in a sealed tube if the following conditions can be met:
   a) The tube is long enough to render the end effects negligible.
   b) The heating effect of the scattered light is eliminated.
   c) The temperature of the confining tube is maintained constant to within 10^{-4} °C.
2. The static pressure rise due to a single laser pulse can probably not be measured if α is as low as 10^{-9} cm^{-1}.
3. The measurement of the acoustic wave generated by a single laser pulse depends on the ussing of the acoustic wave and on an extremely sensitive broad-band detector. It is doubtful that this can be achieved.

4. The measurement of the acoustic signal generated by periodically repeated pulses, whose repetition frequency is matched to that of a resonant cavity, is a possibility that should be investigated further.

5. By use of a phase comparison optical discriminator described in Section 8, values of α in the range 10^{-8} to 10^{-9} cm probably can be measured by interferometric methods.

The situation in the wave length region around 10 microns is quite different from that investigated here. First, Rayleigh scattering is no longer a limiting factor. Second, the absorption in air is considerably higher at 10 microns than it is in certain parts of the visible region. Hence several of the contemplated methods would work. The absorptivity spectrophone was used to measure α in air for the 10.6μ CO_2 laser line. $\alpha = (1.0 \pm 0.2) \times 10^{-6}\ cm^{-1}$.

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Experimental methods proposed for the measurement of the absorption coefficient of light in air are analyzed. The calculations are primarily intended for the wavelength region where, for certain wavelengths, the absorption coefficient to be measured is estimated to be of the order of 10^{-9} cm^{-1}. The effects of scattering, heat conduction, static and acoustic pressure variations are examined and are related to the problem of detecting the signals produced by the absorption of heat from the laser beam. Both pulsed and cw schemes are considered. The measurement of the static pressure rise in a narrow tube appears as a feasible, although marginal method which offers some hope of success. Also an interferometric method appears feasible. Finally the absorption coefficient of the 10.6μ CO_2 laser beam was measured in air, ( ) $\alpha = (1.0 \pm 0.2) \times 10^{-6}$ cm^{-1}.

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